United States Court of Appeals
for the
District of Columbia Circuit

TRANSCRIPT OF RECORD

BRIEF FOR APPELLANT

IN THE

United States Court of Appeals

FOR THE DISTRICT OF COLUMBIA

Appeal No. 8170

EASTMAN KODAK COMPANY,
Appellant,

917

vs.

CONWAY P. COE, Commissioner of Patents,
Appellee.

APPEAL FROM THE JUDGMENT OF THE DISTRICT COURT OF THE UNITED STATES FOR THE DISTRICT OF COLUMBIA

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EASTMAN KODAK COMPANY

Appellant,

vs.

CONWAY P. COE, Commissioner of Patents,

Appellee.

BRIEF FOR APPELLANT

This is an appeal from the Final Judgment (dated Nov. 17, 1941) of the United States District Court for the District of Columbia (Appellant's App. 8) dismissing Appellant's Bill of Complaint which sought the grant of United States Letters Patent containing claims 28 and 29 (Appellant's App. 1) of Bean Application Serial No. 134,903 (Appellant's App. 65).

JURISDICTIONAL STATEMENT

The U. S. District Court for the District of Columbia obtained jurisdiction of this case by virtue of Section 4915 of the Revised Statutes (U. S. C. Title 35, Section 63) under which Appellant's Bill of Complaint (Appellant's App. 1) was filed on Aug. 23, 1941.

This Honorable Court of Appeals has jurisdiction of this Appeal under the Act of February 9, 1893, c. 74, 27 Stat. 434, as amended, and as incorporated in the Code of the District of Columbia, Tit. 17, Sec. 101, the appeal being properly docketed and prosecuted according to the Rules of this Court.

STATEMENT OF CASE

After due proceedings in the U. S. Patent Office, in which the Board of Appeals rejected (Appellant's App. 111) claims 28 and 29 (Appellant's App. 112) of Bean Application Serial No. 134,903 (Appellant's App. 65), Appellant—as assignee of said Bean Application—filed its Bill of Complaint (Appellant's App. 1) against the Commissioner of Patents alleging his refusal to grant Letters Patent containing claims 28 and 29, which are directed to a new chemical compound. The Commissioner duly answered (Appellant's App. 3) alleging said claims 28 and 29 to be unpatentable over certain prior patents which *name* broadly or generically the compounds in question, but which do not describe them or tell how to make them.

Full trial was had in due course, at which Plaintiff took the testimony of several witnesses (Appellant's App. 9 to 64), showing the complete inadequacy of the prior art relied upon by the Commissioner, the detail involved in discovering these new compounds, the date on which Bean made his invention and the commercial success thereof.

The Commissioner's Solicitor cross-examined Plaintiff's witnesses but produced none on his own behalf; the prior art which the Commissioner finally relied upon to anticipate Bean's invention was boiled down to — Wade British Patent No. 350,438 dated June 5, 1931 (Appellant's App. 103), Clifford U. S. Patent No. 2,109,164, filed Oct. 22, 1932 (Appellant's App. 99) and Rogers and Voorhees U. S. Application Serial No. 427,416, filed Feb. 10, 1930 (Appellant's App. 86) later abandoned and replaced by the enlarged Rogers and Voorhees U. S. patent No. 2,084,977, filed March 26, 1937 (Appellant's App. 98) nearly three years after Bean's invention date; the Solicitor also introduced Von Bramer and Ruggles U. S. Patent No. 2,163,639, filed Aug. 17, 1936 (Appellant's App. 116) but it is incompetent because its filing date is too late—some two years after Bean's invention date. The remaining prior art is admittedly merely cumulative, hence we shall not complicate this brief by referring further to it.

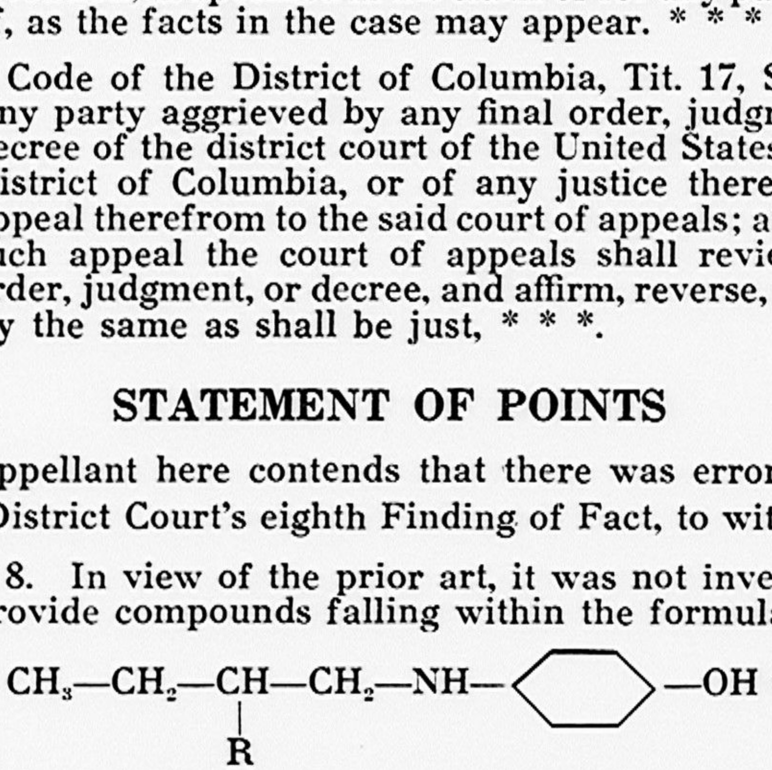
STATUTES

The pertinent statutes upon which the decision of this case depends are as follows:

> Sec. 4886. R. S. (U. S. C., title 35, sec. 31). Any person who has invented or discovered any new and useful art, machine, manufacture, or composition of matter, or any new and useful improvements thereof, * * * not known or used by others in this country, before his invention or discovery thereof, and not patented or described in any printed publication in this or any foreign country, before his invention or discovery thereof * * * may, upon payment of the fees required by law, and other due proceeding had, obtain a patent therefor.
>
> Sec. 4915. R. S. (U. S. C., title 35, sec. 63). Whenever a patent on application is refused by the Board of Appeals * * * the applicant, * * * may have remedy by bill in equity, if filed within six months after such refusal or decision; and the court having cognizance thereof, on notice to adverse parties and other due proceedings had, may adjudge that such applicant is entitled, according to law, to receive a patent for his invention, as specified in his claim or for any part thereof, as the facts in the case may appear. * * *
>
> Code of the District of Columbia, Tit. 17, Sec. 101. Any party aggrieved by any final order, judgment, or decree of the district court of the United States for the District of Columbia, or of any justice thereof, may appeal therefrom to the said court of appeals; and upon such appeal the court of appeals shall review such order, judgment, or decree, and affirm, reverse, or modify the same as shall be just, * * *.

STATEMENT OF POINTS

1. Appellant here contends that there was error in fact in the District Court's eighth Finding of Fact, to wit:

> 8. In view of the prior art, it was not invention to provide compounds falling within the formula:
>
> 
>
> wherein
>
> R is a member from the group consisting of hydrogen and an alkyl group of not more than two carbon atoms.

2. While the District Court's first Conclusion of Law is so entitled, it is believed that under Minnesota Mining and Manufacturing Company v. Coe — App. D. C. —; 125 Fed. (2d) 198, "The common practice of designating findings on invention and patentability, and underlying findings which support them, as conclusions of law is erroneous," and, therefore, that there was error in fact in the District Court's first Conclusion of Law, to wit:

> 1. Claim 29 of the Bean application, Serial No. 134,903, is anticipated by the British patent to Wade, No. 350,438, the patent to Clifford, No. 2,109,164, and the patent to Rogers and Voorhees, No. 2,084,977, and claim 28 thereof is not patentable in view of this prior art.

3. By a parity of reasoning Appellant contends that there was error in fact in the District Court's third Conclusion of Law, to wit:

> 3. The N-(n-primarybutyl)-p-aminophenol of Bean claimed in claim 29 stands anticipated by the British patent to Wade, the patent to Clifford, and the patent to Rogers and Voorhees, regardless of whether this prior art does or does not disclose a process for preparing the compounds, and regardless of whether or not this prior art discloses a process of preparing these compounds which is operative.

SUMMARY OF ARGUMENT

1. A chemical compound is a proper subject for Letters Patent of the United States.

2. In 1934 Bean independently discovered and prepared N-(n-primary butyl)-p-aminophenol; in 1936 he discovered N-(2-ethyl-n-primary butyl)-p-aminophenol; in late 1936 and early 1937 he determined the efficacy of these compounds as gasoline antioxidants.

3. The only evidence relied upon by the Commissioner of Patents is three prior patents which do no more than list *names* of theoretical compounds. This does not constitute evidence of their *existence*.

4. Plaintiff's witnesses have testified that the *mere name* of an unknown chemical compound, whether generic or specific, is not a sufficient description to depict its properties or preparation and hence to permit its utilization.

5. Defendant's Evidence.

   A. The Wade British patent No. 350,438 discloses a mere generic *name*.

   B. The abandoned Rogers and Voorhees application Serial No. 427,416 discloses a mere generic *name*.

   C. The Clifford U. S. patent No. 2,109,164 discloses a mere *name*, somewhat more specifically than the Wade British patent or the Rogers and Voorhees application.

   D. The Von Bramer and Ruggles U. S. patent No. 2,163,639 is not a proper reference against N-(n-primary butyl)-p-aminophenol (the only one of Bean's compounds which the patent purports to disclose), since it was not filed until August 17, 1936, long after Bean's invention.

6. Plaintiff's Evidence.

   A. Testimony of Bean and Vaughn show that prepara- of Bean's compounds was not obvious from the prior art.

   B. Testimony of Tanghe clearly shows that Bean's compounds were not even suggested by the prior art.

   C. Testimony of Davy clearly shows that the prior art contains no anticipation of the applicant's compounds and that there was, at the time of the prior patents, no literature to which one could turn to obtain any description of any of the compounds claimed by Bean.

   D. Commercial success of Bean's N-(n-primary butyl)-p-aminophenol as a gasoline antioxidant was clearly established by Dryer's testimony, and while such success alone is not evidence of invention, it is—coupled with a six-year desire by the industry for efficacious antioxidants—persuasive that Bean's compounds existed in no more than name prior to Bean's invention.

7. The District Court's eighth Finding of Fact that Bean's compounds lacked invention is clearly erroneous and inconsistent with the evidence. The same is true of the District Court's first and third Conclusions of Law (treated as Findings of Fact) in which the Court holds that the three principal prior art references constitute anticipation of Bean's claim 29.

8. No evidence favors the Commissioner. But, the mass of evidence presented by the Plaintiff is substantial enough to convince a reasonable mind that the *mere name* of a generic group of compounds is not evidence of the existence of any of those compounds, but merely a suggestion or speculation that such compounds may some day be provided.

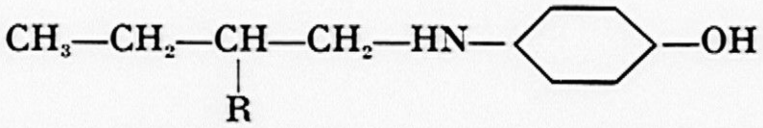
9. The decisions of the District Court of the District of Columbia, upon the issues here presented, are indeed conflicting and need to be authoritatively clarified.

ARGUMENT

In 1934 Bean invented N-(n-primarybutyl)-p-aminophenol which is the chemical compound defined by claim 29 of his application Serial No. 134,903 which was filed on April 3, 1937; in 1936 he invented N-(2-ethyl-n-primarybutyl)-p-aminophenol, completing the group of chemical compounds embraced in claim 28 of his application; also in 1936 he discovered that these chemical compounds were very effective as antioxidants for preventing gum formation in gasoline (See Finding of Fact No. 7 — Appellant's App. 7).

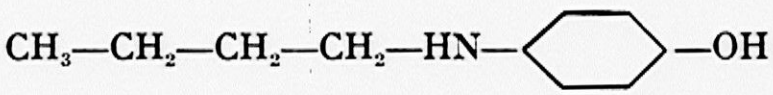
These two claims, the only ones involved in the Civil Action below and in this appeal are as follows:

28. A compound having the formula:



wherein R is a member from the group consisting of hydrogen and an alkyl group of not more than two carbon atoms.

29. A compound having the formula:



Claim 28 claims a group of chemical compounds having 4, 5, or 6 carbon atoms in the alkyl group, attached to the nitrogen atom of the compound. Claim 29 defines the specific chemical compound in which n-primarybutyl constitutes the alkyl group attached to the nitrogen.

That a chemical compound is a proper subject for Letters Patent of the United States is provided by R. S. 4886 which provides that anyone who has invented any new *composition of matter* may obtain a patent therefor, upon due proceedings had. Bean's claimed chemical compounds constitute invention because they were new at the time of his discovery thereof and because they were not previously patented or *described* in any printed publication in this or any foreign country. No one has questioned the utility of Bean's new compounds. The record is replete with commendation. Accordingly claims 28 and 29 here on appeal, constitute a proper statutory form of invention.

But the Commissioner of Patents contends and the District Court has held that Bean's claims 28 and 29 are anticipated by the prior art or are not patentable in view of this prior art. This we will show is erroneous.

We appreciate fully that in view of such decisions by this Court as Abbott v. Coe, 71 App. D. C. 195; 109 Fed. (2d) 449, Appellant's burden is to convince this Court that the District Court's Findings are not consistent with the evidence.

The Commissioner's Evidence

The only evidence presented by the Commissioner of Patents, of any consequence whatsoever, is the Wade British patent No. 350,438 (Appellant's App. 103), Clifford U. S. Patent No. 2,109,164 (Appellant's App. 99), and the Rogers and Voorhees Application Serial No. 427,416 (Appellant's App.

86) which patents and application do no more than to *list* the *names* of certain compounds which at the time of those patents were wholly theoretical compounds—no properties thereof or method of preparation being given.

We submit that it does not constitute evidence of the existence of a chemical compound merely because the *name* thereof (whether specifically or generically) appears in print in a prior publication. Jules Verne visualized the submarine but it remained for others to invent it and give it to the Navy.

Plaintiff's witnesses repeatedly testified and emphasized the fact that the mere *name* of a chemical compound appearing in print does no more than to indicate to the chemist the number, kind and approximate grouping of the various atoms going to make up the chemical compound but this does not tell one any of the physical properties or any of the chemical properties of the compound in question.

For instance Bean testified under questioning by the Court (Appellant's App. 24):

> "By the Court:
>
> Q. In this Clifford patent on page 2, specifications line 8, isn't there a mention of N-butyl-p-aminophenol?
>
> A. Clifford? Page 2?
>
> Q. Clifford, page 2, line 8.
>
> A. Second column. There is mention of nitrogen butyl para-aminophenol. That doesn't tell me what the compound is at all from my knowledge of organic chemistry. I have no knowledge of what kind of butyl group that was, what its properties were, or anything else."

Bean further testified, under recross examination by defendant's counsel (Appellant's App. 25):

> "Q. You have used the same language as Clifford?
>
> A. I have used, to me, what you would call a shortened terminology, and my use of that is based on my preparation. I know what I've got, and I have melting points of it, and it is a very definite thing to me, and it

is no more than if I sent it down to them marked with 'X'. I was just interested in the results.

> The Court: The question was whether your description in those papers was the same as in the Clifford one. That was your question?
>
> Mr. Moore: That is the question, your Honor.

By the Court:

Q. Will you answer the question?

A. Yes.

Q. It is the same?

A. But the meaning back of it, to me is: To me, I prepared this compound on the basis of synthesis, and I know the structure of it, and I refer to that, and I can't see where the name has any bearing on it."

Bean further testified under redirect examination by counsel for plaintiff (Appellant's App. 27):

> "Q. Just one point. Back of this name business you had a method of preparation, did you not Doctor?
>
> A. Yes, sir.
>
> Q. And that method of preparation was described in connection with your mention of nitrogen-butyl-para-aminophenol in a number of cases, was it?
>
> A. Yes, sir.
>
> Q. And would that, to you, or to anyone skilled in the art, also knowing the method, mean a broad compound, or would that mean the compound that resulted from the synthesis?
>
> A. It would mean the compound from the synthesis."

Dr. Davy, an expert organic chemist, testified as to the meaning of a mere *name* of a chemical compound as follows (Appellant's App. 38 and 39):

> "Q. When you say there are 44 possibilities there and that the name 'butyl aminophenol' may mean anyone or all 44 of them, does that name tell you anything about the properties of the compounds?
>
> A. It tells a certain amount to one who is familiar with the aminophenols and their chemical properties.

> That is, that a part of the group would be expected to be soluble or to form salts with strong alkalis or with strong acids due to the presence of the phenolic hydroxyl and the amino group; *but as regards the physical properties or other properties it conveys nothing.**
>
> Q. Does it tell you anything about the color of the compound.
>
> A. No. I think it might be colorless. But there is nothing definite; no.
>
> Q. Does it tell you anything about its melting point or boiling point.
>
> A. No.
>
> Q. Does that name tell you anything about its properties toward other chemicals or compounds other than the broad fact that it is a phenol.
>
> A. It says that it will possess in general the chemical reactivity of a phenol and an aminophenol, but nothing such as derivatives of the amino group and the hydroxyl group.
>
> Q. Does the name 'butyl aminophenol' tell you, speaking as of the date in 1930, how to prepare the compound?
>
> A. Not at all.
>
> Q. In 1930 was there known in the literature any method of preparation of any butyl aminophenol?
>
> A. No.
>
> Q. Did you yourself know any in 1930?
>
> A. No."

Thus in 1930 (when Rogers and Voorhees filed), in 1931 (when Wade issued), and in 1932 (when Clifford filed) and in 1934 (when Bean discovered these compounds) N-(n-primarybutyl)-p-aminophenol was not a compound which one could purchase over the counter at any corner drugstore or at even the best stocked chemical supply house. It was not even a compound which, in so far as the Commissioner of Patents has proven, one could obtain from the leading chemical authorities of the world. In fact in so far as the record shows, one could demand and plead of the entire chemical industry that the compound be supplied and it would not be forthcoming, and one could demand and plead that the properties and method of preparation of the compound be elucidated—without avail, yet the Commissioner of Patents would have the Court believe that the mere printed *name* of the chemical compound is evidence of its existence.

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* Italics our emphasis throughout this brief unless otherwise noted.

A — The Wade British Patent No. 350,438

All this patent discloses is the mere recitation that

> "para- and ortho-methyl-amino-phenol, dimethyl-aminophenol, *butyl,* isopropyl and benzyl *amino-phenols,* dihydroxy-diphenyl-amine, and the like"

will stabilize gasoline against gum formation but this does not constitute evidence of the *existence* of such compounds in 1931. Dr. Davy's testimony before the District Court showed that there were forty-four possible compounds under Wade's broad language that "butyl * * aminophenols" could be used as gasoline stabilizers. What in these three mere words constitutes evidence that the compounds existed, what were their properties, and how could they be prepared? Certainly the three words do not constitute evidence which anticipates Bean's discovery of N-(n-primarybutyl)-p-aminophenol—one of the forty-four compounds embraced in Wade's broad speculation.

B — The Abandoned Rogers and Voorhees Application Serial No. 427,416

The use by the Commissioner of this application in this proceeding is permissible only because it constitutes the parent of the application which matured into Patent No. 2,084,977. Conclusion of Law No. 4 clearly holds that the Rogers and Voorhees Patent No. 2,084,977 is good only for that which was disclosed in the parent application Serial No. 427,416. Accordingly this bit of evidence is simplified by referring only to the parent application, which discloses the mere recitation that—

> "the aminophenols, para- and ortho-methyl aminophenol, dimethyl aminophenol, hydroxyazobenzine, *butyl*, isopropyl and benzyl *aminophenols*, dihydroxy diphenylamine, paraphenetidine, amino naphthol and the like"

may be used to stabilize gasoline. But this is no description of "butyl * * aminophenols" or anything about the properties or anything about the process for preparing them. Just as in the case of the British patent to Wade, the mere *recitation* of the words "butyl aminophenols" is no evidence of the *existence* of the forty-four compounds embraced in this language.

C — Clifford U. S. Patent No. 2,109,164

This patent is a remarkable compilation of some 150 compounds and groups of compounds which the patentee would lead the public to believe, existed and could be used principally as rubber anti-oxidants although their possible utility as gasoline anti-oxidants was also adverted to. Among this long list of compounds Clifford refers to "N-butyl-p-aminophenol." Even with this somewhat more specific group of such phenols, this terminology embraces four possible compounds. Which one is the public to believe that Clifford had in mind; certainly not all of them. But Clifford goes a little further in his speculations than do the other two pieces of prior art relied upon by the Commissioner in that Clifford postulates on page 3, column 2, lines 17 to 20, that "N-butyl p-amino phenol can be prepared by reacting butyl amine and p-chloro phenol in an autoclave." (Appellant's App. 101.) That is the only clue that Clifford gives as to the *existence* of the compounds.

In an effort to ascertain whether Clifford had contributed something actual to the art, Plaintiff's witnesses Vaughn and Davy independently made every effort to produce a N-butyl p-amino phenol by Clifford's method but were wholly unable to obtain any such product (See Vaughn's testimony at Appellant's App. 33 and Davy's testimony at Appellant's App. 47). These compounds simply did not exist before Bean's invention thereof.

The fifth Finding of Fact states that "plaintiff did not prove that the method referred to on page 2, column 2, lines 60 to 75, of that patent is inoperative to prepare that compound." This is wholly beside the point inasmuch as Clifford states at line 60 (Appellant's App. 100) that "In general, methods for preparing the antioxidants described above will suggest themselves." Prior to that point Clifford has named some 150 compounds and groups of compounds of the most complicated theoretical type. Who is it that is going to suggest the method of preparing these 150 compounds? Certainly if Clifford does not suggest the method, the Clifford patent does not constitute evidence. The compound, the preparation of which Clifford does describe at this point, is entirely different from the N-butyl p-amino phenols, one of which is here in issue. Certainly it is not the Plaintiff's task to prove such a method inoperative but the Commissioner's task to prove that such a method is operative to produce the compounds in issue.

Clearly Clifford is nothing more than a speculative list of *names* and does not constitute evidence of the *existence* of a single one of them.

D — The Von Bramer and Ruggles U. S. Patent No. 2,163,639

Through some abortive reasoning the Commissioner's Solicitor attempted at the trial to invoke the teachings of the Von Bramer and Ruggles patent to bolster the Clifford patent. This whole attempt obviously fails completely inasmuch as the Von Bramer and Ruggles application was not filed until Aug. 17, 1936, more than two years after Bean fully reduced his invention to practice. Reference to this Von Bramer and Ruggles patent does not appear in the Findings of Fact or Conclusions of Law and we, of course, object to the Appellee's burdening the record with this patent or any reference thereto.*

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* Incidentally, the rather unusual size of Appellant's Appendix is due to the Commissioner's Solicitor's desire to have included in the Printed Record on Appeal a number of pieces of evidence which we did not believe pertinent to the issues involved in this appeal.

Thus the Commissioner's evidence boils down to nothing but three principal pieces of prior art—Rogers et al 1930 application, Wade British patent and Clifford U. S. patent (and a few cumulative ones which merit no discussion) all of which go no further than merely to *name* "butyl aminophenols" or "N-butyl-p-amino phenol" without one scintilla of proof that such compounds *existed* at the time of those publications. If a publication refers to ethyl alcohol, formaldehyde, acetone or any such well known chemical compound, no description or process of preparing it need accompany such name as the compound has long been well known to the art and the mere mention of the name conveys to the chemist its complete physical and chemical properties, but when a publication referred to a butyl aminophenol in 1930, 1931 or 1932 it meant to no chemist any more than *a mere name*. It did not tell the chemist whether the compound was liquid or solid; if solid, what was its crystalline structure; what was its melting point or boiling point, what was its reactivity toward known chemicals and—above all—how to obtain the compound. The mere name simply is not *evidence* of the *existence* of the compound.

Accordingly there is really no basis whatsoever in the evidence presented by the Commissioner of Patents for the Finding that claim 29 of the Bean application is anticipated by Rogers and Voorhees, Wade or Clifford, or that Bean's claim 28 is not patentable over this prior art. All the more so does the evidence fail to support any finding that Bean's claim 29 is anticipated regardless of whether the prior art discloses a process for preparing the compounds. Certainly that which did not *exist* (except in theory and speculation) cannot anticipate.

Plaintiff's Evidence

Not only did Bean testify fully as to the meticulous care and careful deductions necessary to produce N-(n-primary-butyl)-p-aminophenol (covered by his claim 29) and the properties of such compound (Appellant's App. 10-11) but he testified that none of the prior art gave him any help in arriving at this compound or any of the other small group of compounds covered by his claim 28.

Furthermore, Vaughn testified as to how he collaborated with Bean in carrying out Bean's ideas and how it was only with meticulous and detailed care that one could arrive at the preparation of the compounds claimed by Bean. Certainly there is not one whit of testimony by either of these witnesses to show that the mere *naming* of a compound gave sufficient information to reproduce it. Not content with the testimony of Bean and his associate, Plaintiff had such skilled chemists as Tanghe, Davy, and Dryer testify as to their views upon and experience with the situation.

Tanghe's testimony under direct questioning by the District Court is very enlightening. Tanghe worked with this general type of chemistry, was familiar with all of the starting materials employed by Bean in arriving at his invention and had considerable experience with related reactions. After corroborating Bean's preparation of compounds covered by claims 28 and 29, the following colloquy occurred between the Court and the witness (Appellant's App. 29):

> "Q. Just what, in your opinion, was the discovery that was made by Dr. Bean in getting this result which is in Exhibit 5?
>
> A. In my opinion, *he was the first person to have this butyl para aminophenol* in question.
>
> Q. How was it that it was something new that was never known before? What did he do that nobody else had ever done before?
>
> A. He put these various chemicals together under favorable conditions for the formation of the compound and isolated it as a fairly pure substance.
>
> Q. What chemical did he put in that no one else had ever put in?
>
> A. He used a method involving para aminophenol with butyraldehyde. Previous mentions of the compound are not very definite as to what was placed in

> the reaction to obtain the compound or whether it had been obtained, even. *I think his work shows conclusively that he actually had this compound,* and that point can hardly be questioned from the way in which the synthesis was carried out.
>
> Q. Did not the prior art suggest that he might have obtained this resultant?
>
> A. By carrying out the reaction as Dr. Bean has?
>
> Q. Yes; or like Clifford suggested, or like any other expert chemist might suggest.
>
> A. *No; I do not think the prior art did.*
>
> Q. Can you tell the Court just what he did that nobody else had done or suggested before?
>
> A. He actually carried out the synthesis on this compound in a way which, so far as I know, had not been suggested before.
>
> Q. What was that way?
>
> A. By condensing an aldehyde with para aminophenol and reducing the compound—reducing the intermediate compound."

Thus the evidence is clear that the prior art such as Clifford, Wade, and Rogers and Voorhees did not suggest or describe the compounds claimed by Bean or how to prepare them and the evidence is, therefore, such that the District Court's eighth Finding of Fact is clearly erroneous and inconsistent with the evidence. The same is true of the District Court's first and third Conclusions of Law (treated as Findings of Fact) in which the Court holds the three principal prior art references relied upon by the Commissioner constitute anticipation for Bean's claim 29. As aptly said by the Third Circuit Court of Appeals in Skelly Oil Co. v. Universal Oil Products Co., 31 Fed. (2d) 427, at 431—

> "A patent relied upon as an anticipation must itself speak. Its specification must give in substance the same knowledge and the same directions as the specification of the patent in suit. Otto v. Linford, 46 L. T. (N. S.) 35, 44. It is not enough to prove that a method or apparatus described in an earlier specification can be made to produce this or that result. Flour Oxidiz-

> ing Co. v. Carr & Co., 35 R. P. C. 457. A singularly sensible test of the rule of anticipation is given in British Thomson-Houston Co. v. Metropolitan Vickers Electrical Co., 45 R. P. C. 22, by asking the question — *'Would a man who was grappling with the problem solved by the patent attacked, and having no knowledge of that patent, if he had had the alleged anticipation in his hand, have said: "That gives me what I wish"?'* ".

Again in the case of American Stainless Steel Company v. Ludlum Steel Company, 290 Fed. 103, Justice Hough stated:

> ". . . but it requires more than *prophecy* of what may be done or than declaration of what ought to be accomplished, to make a good patent reference, not to speak of anticipation."

Accordingly following the well-known principals of law there is not one scintilla of real evidence (nothing more than a mere name) that Bean's claims 28 and 29 are anticipated. Accordingly the District Court's Conclusions Nos. 1 and 3 are erroneous because entirely inconsistent with the evidence.

Testimony of the same character was adduced from the witness, Dr. Davy. He was an experienced chemist working for years with this type of chemistry and this general type of chemical reaction. At the request of attorneys for the Plaintiff he thoroughly tested out the process alleged by Clifford to produce N-butyl p-amino phenol and found it wholly inoperative. The proof of this inoperativeness is sustained by the District Court's fifth Finding of Fact. As to the Commissioner's suggestion that the method referred to on page 2, column 2, of the Clifford patent might produce N-butyl p-aminophenol, Dr. Davy clearly showed the impossibility of such a suggestion (Appellant's App. 48). Dr. Bean further denied (Appellant's App. 23) that any help could be obtained from the Von Bramer and Ruggles patent to bolster the Clifford patent even though the Von Bramer and Ruggles patent might be regarded as competent evidence (which we assert that it is not).

Dr. Davy gave considerable aid to the District Court in showing that forty-four chemical compounds were possible from the mere name—butyl aminophenol (referred to by Wade and by Rogers and Voorhees—Appellant's App. 37-38). He further showed that even taking the Clifford patent at its best, it contained no more than the name N-butyl p-aminophenol which name included four possible compounds (Appellant's App. 36-54).

Dr. Davy further showed that prior to Bean's invention (namely during the Clifford, Wade, and Rogers and Voorhees era) there was no literature, no textbook to which one could turn to obtain any description of any of the compounds claimed by Bean (Appellant's App. 39). Thus the *mere name* of a compound does not give that compound to the chemist. Obviously had there been any such literature or any such textbook, the Commissioner most certainly would have drawn the Court's attention thereto. As a matter of fact had such information existed, this case would never have been tried.

Dr. Davy then testified that even with the exact name of any compound, such as those claimed by Bean, the chemist would not know how to prepare it or where to obtain the compound (Appellant's App. 42-43). True he could try this reaction and that reaction in an effort to arrive at something having the same name and which the chemist might deduct was intended by the name but that is no description of the compound and virtually begs the question as that is the essence of Bean's invention, namely the provision of the very compounds claimed by him.

Thus we have unassailed evidence that the mere name of these compounds did not give them to the industry but that it required most ingenious chemical perception to obtain the compounds and provide them for the industry. Is this not complete evidence that Bean performed an invention in providing these compounds? Certainly it is clear that there is no evidence to support the District Court's eighth Finding of Fact holding that these compounds are not invention.

We realize that proof of commercial success is not evidence with which—alone—it is inconsistent to find no invention. However, when such commercial success is coupled with a six-year desire by the industry for the very compounds in question, it certainly is most persuasive evidence that the compounds existed in no more than *name* during that entire six years or at least during that part of it prior to Bean's invention.

It is too well known to require proof that the Standard Oil Company sells millions of gallons of gasoline annually. It is also clear from the prior art that gasoline antioxidants are most desirable. It is well nigh inconceivable that the Standard Oil Company should have had the Wade British patent No. 350,438 on file since 1930 and granted since 1931 and still not have utilized and made available to the public any butyl aminophenol, had these compounds represented anything more than a mere generic name contained in this British patent. The pride of human accomplishment and the desire for gain by virtue of improvement are too strong to have permitted any of Bean's claimed compounds to have existed since 1930 and not have been widely known and fully described in the chemical literature and in the industry.

The witness Dryer with more than a decade of experience in the business of furnishing antioxidants to the gasoline industry testified unequivocally to the fact that prior to Bean's invention, no butyl aminophenol was known to the petroleum industry (Appellant's App. 62) that the gasoline industry had been employing an entirely different chemical compound (benzyl p-aminophenol) as an antioxidant and that it was not until N-(n-primarybutyl)-p-aminophenol was presented to Dryer's organization "on a silver platter" by Bean's assignee that the gasoline industry ever knew of the existence of the compound. It should be plain that the highly competitive gasoline industry of the last decade does not employ inferior compositions (benzyl aminophenol) when much more efficient compounds are "staring them in the face" and are described in their own patents. Commercial success of Bean's invention is, therefore, highly persuasive evidence of the fact that the Wade British patent was no more than a speculation as to the possible *future* existence of any of the butyl aminophenols.

As aptly stated by the Second Circuit Court of Appeals in the case of Westinghouse Air-Brake Company v. Great Northern Railway Co., 88 Fed. 258—

> "The prophetical suggestions in English patents of what can be done, when no one has ever tested by actual and hard experience and under the stress of competition the truth of these suggestions, or the practical difficulties in the way of their accomplishment, or even whether the suggestions are feasible, do not carry conviction of the truth of these frequent and vague statements."

No Evidence Favors the Commissioner

Thus the Commissioner has presented no evidence save for a *name*, and an indefinite and generic name at that. The name butyl aminophenols used by Rogers and Voorhees and by the Wade British patent covers forty-four possible compounds. Even Clifford's more specific name, N-butyl p-aminophenol covers four possible compounds. The sole question of this case is whether such meager speculative evidence is sufficient to support the District Court's Findings of "no invention" and "anticipation" or, to state it otherwise, whether the District Court's Findings were inconsistent with the mass of contrary evidence presented by the Plaintiff.

With all due respect to the District Court and its willingness to receive our evidence and hear our presentation of the case, we do not believe that it fully appreciated the issue which we presented for decision, for the reason that after presentation of our evidence and full argument of the case following the presentation, the following colloquy between the Court and counsel for Plaintiff occurred (Typewritten Transcript of Proceedings, Vol. 3, pp. 164, 165):

> "The Court. Suppose an expert chemist were told the substance which is in this bottle (indicating): if he were asked to write a formula, could he do it?

> Mr. Mayne. Yes, sir.
>
> The Court. Just as it is on the blackboard?
>
> Mr. Mayne. Yes, sir.
>
> The Court. Then he would know that that formula could be gotten together in a crucible, in a chemical laboratory in different ways, with heat applied in different degrees, and you say he might get four different compounds of the same nature, four varieties. Is that right?
>
> Mr. Mayne. That is just the point, your Honor. Clifford suggested a method, but it would not work. Clifford did not have that compound.
>
> The Court: I am not talking about the method; I am just saying that you could write that formula and put those chemicals together and get a compound called nitrogen butyl aminophenol?
>
> Mr. Mayne. That is just the point. You cannot put these together. We cannot go and pick up a phenol and pick up a butyl radical and add them together."

Thus it would appear that "up to the last" the Honorable District Court was under the impression that a chemical compound, being capable of being represented by a formula, is like a machine constituted of various parts capable of being assembled at will and that all that is necessary in order to assemble such a chemical compound (having the theoretical formula before one) is to take from the shelf of the chemical laboratory, a benzene ring, a hydroxy group, an amine group, an alkyl radical, and a bunch of carbon and hydrogen atoms (which would be required by the formula of Bean's claimed compounds), put them "together in a crucible, in a chemical laboratory in different ways, with heat applied in different degrees" and obtain the compound. Such is (as clearly shown by the evidence) far from the real procedure required to obtain Bean's compounds and is the frequent error of the lay mind—those not skilled in the intricacies and difficulties of preparing chemical compounds—and we naturally do not expect a District Court to be skilled in the art of chemistry.

But we do submit that it is entirely inconsistent with the clear-cut evidence proving that the mere publication of the name of a chemical compound does not constitute evidence of the existence of the compound, to hold that there is no invention in providing that compound and giving it to the industry.

Thus the Commissioner has proven no more than the existence of the mere generic *name* of the compounds claimed by Bean and the Honorable District Court was possessed to the last with the impression that a chemical compound results from the mere fitting together of the various atoms at will in accordance with a formula. On the other hand, Bean has proven that he independently discovered and actually produced the compounds which he has claimed. He and three other experts have testified in one vein or another, that the mere name of those compounds did not prove their existence and that it was left for Bean to discover them and provide them. How can it in any way be consistent with this evidence for the District Court to find that these compounds—claimed by Bean—are anticipated by the prior art or that it is no invention or not patentable to provide such compounds?

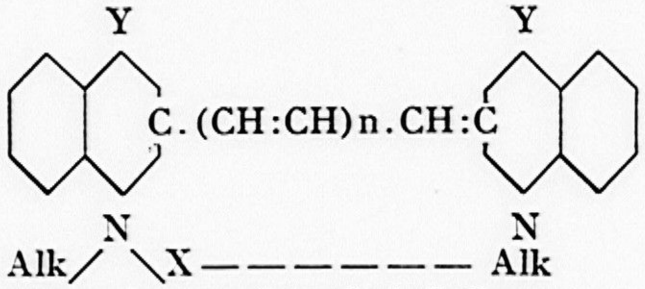
While we believe that this is the first time this Honorable Court of Appeals has been presented with this question in connection with a chemical compound, this Court has clearly enunciated in the case of Minnesota Mining and Manufacturing Company v. Coe, 73 App. D. C. 146; 118 Fed. (2d) 593, that "The question of invention is the question whether 'some uncommon talent' is shown." Certainly with the industry having the name "butyl aminophenols" before it for six years and with the experts being unable to prepare such compounds after the name was before them and with the industry exercising its "long felt want" for N-(n-primarybutyl)-p-aminophenol promptly upon Bean's presentation of the compound to the industry, it is most persuasive that "some uncommon talent" was necessary to provide this compound.

Again in this same Minnesota Mining and Manufacturing Co. v. Coe case this Court stated that "Substantial evidence is 'such relevant evidence as a reasonable mind might accept as adequate to support a conclusion.'" Certainly the mass of evidence presented by Plaintiff is substantial enough to convince a reasonable mind that the *mere name* of a generic group of compounds is *not* evidence of the *existence* of any of those compounds but merely a suggestion or prediction that such compounds may some day be provided.

Accordingly it seems clear that all "Three Points" urged by Appellant should be decided in Appellant's favor and that it should be held that it was *inconsistent* with the evidence for the District Court to hold that there was no invention for Bean to provide the claimed compounds or that the claimed compounds were anticipated by the Rogers and Voorhees application, the Wade British patent or the Clifford U. S. patent.

Conflict of Decisions

Our case would not be fairly presented if we did not call to this Court's attention the conflict upon this subject which exists in the decisions of the District Court of the District of Columbia. We have already familiarized the Court with the decisions and holdings of Justice O'Donoghue in the instant case. But the District Court of this jurisdiction has not always maintained that view. In the case of Eastman Kodak Company v. Coe, 40 Fed. Supp. 891, the Commissioner of Patents had rejected Applicant's claims to a seleno tricarbocyanine dye on the ground that the prior art had published a generic formula with instructions that various atoms could be substituted in that generic formula at various points therein to give a formula which was identical with that of the dye claimed by the Applicant in that case. In other words the prior art portrayed no more than the name and formula of the dye. After full presentation of evidence Justice Bailey held that—

> "The chief ground upon which the Board of Appeals of the Patent Office denied the allowance of the claims sought by the plaintiff was that the product claimed was anticipated by an article described in a publication written by Konig (Konig, 'Berichte,' 57, page 685). Konig gave a general formula for certain dye stuffs as follows:
>
> 
>
>
> wherein n may have values 0, 1, 2, 3, etc.
>
> He also stated that instead of Y a cyclo-seleno group might occur.
>
> In the plaintiff's product Selenium takes the place of Y and n equals 3.
>
> Konig does not state that he has produced such a product as the plaintiff's but *in effect states that such a product may exist as a dye.*
>
> "I think that the production of a new chemical compound, *even though some one has stated that such a compound may exist* is an invention within the meaning of the patent laws and that the plaintiff is entitled to the relief sought."

We submit that the instant case is no different in principle from Eastman Kodak Company v. Coe above referred to. Certainly the decision in the previous case throws serious doubt upon the propriety of the District Court's decision and holding in the instant case. We have felt, therefore, that the instant case should be reviewed by this Honorable Court and consequently have gone to more than the usual extent in presenting the facts of the instant case, bearing upon this point.

In fact the "weight of authority" in the District Court, District of Columbia, would appear to be in Plaintiff-Appellant's favor upon the issues here involved for the reasons pointed out in Nye v. Coe, — Fed. (2d) — (53 USPQ 663), which was an R. S. 4915 case decided on April 13, 1942, by Judge

McDuffie. There, as here, a nebulous disclosure in a British patent was the anticipation alleged by the Commissioner of Patents. Judge McDuffie stated as follows:

> "Upon a casual reading of the modification of the preferred embodiment of the British patent 371,340, the language impresses one as an anticipation of the plaintiff's claim, but upon a careful study and analysis of that language, together with Figure 4 of the British patent referred to, one becomes more and more convinced that *the writer of the modification was in a field of speculation or prophesy.* It becomes more and more difficult, and indeed impossible to read in lines 86 to 93 a full and clear disclosure of the Nye invention defined by claims 14, 16, 20, 21 and 22, as to both structure and contemplated functional utility such as to warrant a denial of plaintiff's right to these claims.
>
> It is not difficult to understand how an examiner could read in the modification of the British patent the anticipation claimed for it, after actually having before him the plaintiff's invention. This, however, would mean the exercise of 'hindsight' as that rather expressive word was used by Mr. Justice Stephens in the case of Becket v. Coe, 69 App. D. C. 51, 98 F. 2d 332, 336 (38 USPQ 26, 30), in which he said:
>
>> 'With the Becket application before us it is possible to look back upon the Hadfield (French) and Commentry (French) patents and to say that they suggested what he found, but such hindsight should not, we think, defeat his application. Hadfield and Commentry seem neither to have sought nor to have found an alloy which is both stainless and deep-drawing, and though it be true that the scope of their ranges embraces the claims of Becket, this speaks of the *extent of their desire* rather than of the breadth of their discoveries.'
>
> * * *
>
> The court is not unmindful of the rule that a patentee need not describe in minutest detail, make experiments, and accompany his descriptions with sketches or drawings, in order that his disclosure may be an anticipation, yet the rule is also clear that before a disclosure can be held to be an anticipation, its language or claims must be so certain and descriptive, that one skilled in the art or one who has been termed a journeyman or an artisan, can visualize therefrom, *specifically, definitely and in detail,* the type of device or construc-

> tion that was in the mind of the patentee who described it.
>
> * * *
>
> Again in Davies v. Coe, 65 App. D. C. 345, 83 F. 2d 602, 603 (29 USPQ 107, 108), the Court restated the rule:
>
> 'The disclosure of a foreign patent is to be measured not by what may be made out of it, but what is clearly and definitely expressed in it. In re Ek, 57 App. D. C. 203, 19 F. 2d 677; Carson v. American Smelting & Refining Co., 4 F. 2d 463.'"

Judge McDuffie then held that the British patent did not teach the invention of the plaintiff Nye and that the plaintiff was entitled to the claims in issue.

One other decision also "enters into the picture" namely, In Re Von Bramer and Ruggles, 127 Fed. (2d) 149, recently decided by the U. S. Court of Customs and Patent Appeals. In that case the claims were directed to gasoline stabilized with an aminophenol (not the aminophenol claimed in the instant case). The Customs Court held virtually that since the prior art had suggested the use of such aminophenols as stabilizers for gasoline, Von Bramer and Ruggles had performed no invention in so stabilizing gasoline. That is far different from the issue here involved which issue relates to new chemical compounds as distinguished from gasoline *containing* compounds already suggested for that purpose.

As a matter of fact the instant appellant had appealed a similar issue to this Honorable Court in the related appeal No. 8169 but withdrew that appeal upon appearance of the decision by the Court of Customs and Patent Appeals in In Re Von Bramer and Ruggles. On the other hand, the instant case represents a separate and more basic issue.

Accordingly we submit that the judgment of the District Court in the instant case should be reversed.

Respectfully submitted,
CLARENCE M. FISHER,
Attorney for Appellant.

NEWTON M. PERRINS,
DANIEL I. MAYNE,
Of Counsel.

July 31, 1942.

APPENDIX TO APPELLANT'S BRIEF

IN THE

United States Court of Appeals

FOR THE DISTRICT OF COLUMBIA

Appeal No. 8170

EASTMAN KODAK COMPANY,
Appellant,

vs.

CONWAY P. COE, Commissioner of Patents,
Appellee.

APPEAL FROM THE JUDGMENT OF THE DISTRICT COURT OF THE UNITED STATES FOR THE DISTRICT OF COLUMBIA

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APPENDIX.

Complaint for Issuance of United States Letters Patent.

IN THE DISTRICT COURT OF THE UNITED STATES

FOR THE DISTRICT OF COLUMBIA

CIVIL ACTION No. 12,618

EASTMAN KODAK COMPANY
343 State Street, Rochester, New York
Plaintiff

VS.

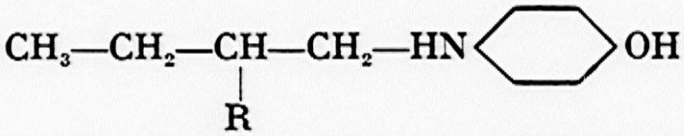
CONWAY P. COE, Commissioner of Patents,
Washington, D. C.
Defendant

I. The jurisdiction of this Court depends upon the Patent Laws of the United States, this complaint being filed under Section 4915, Revised Statutes (Comp. Stats. 9460; U. S. C. Title 35, Section 63), as amended by Act of March 2, 1927.

II. The plaintiff, Eastman Kodak Company, is by an assignment in writing executed on the 29th day of March, 1937, and recorded in the United States Patent Office on August 5, 1941, in Liber Q188, Page 158, of the Transfers of Patents, the sole owner of the entire right, title and interest in an application for United States Letters Patent of Frederic R. Bean, for improvements in N-SUBSTITUTED AMINOPHENOLS AND MOTOR FUELS STABILIZED THEREWITH, filed in the United States Patent Office on April 3, 1937, Serial No. 134,903, and the improvements disclosed and claimed therein.

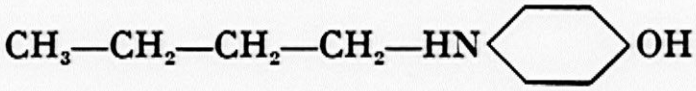
III. The Board of Appeals of the United States Patent Office has refused to allow two product claims of said Frederic R. Bean application, Serial No. 134,903, which claims are as follows:

28. A compound having the formula:



wherein R is a member from the group consisting of hydrogen and an alkyl group of not more than two carbon atoms.

29. A compound having the formula:



IV. The defendant, Conway P. Coe, Commissioner of Patents of the United States, by the aforesaid decision of the Board of Appeals of the United States Patent Office has refused and still refuses to grant Letters Patent of the United States to the plaintiff, Eastman Kodak Company, as assignee of said Frederic R. Bean, on his said application, Serial No. 134,903, containing the two claims recited in Paragraph III hereof.

V. The said decision of the Board of Appeals was rendered on or about the 7th day of May, 1941 (supplemented, upon request for reconsideration, by a decision rendered on or about the 1st day of July, 1941), and within six months last past, and no appeal has been taken from said decision of the Board of Appeals to the Court of Customs and Patent Appeals, and no such appeal is pending, or has been decided.

WHEREFORE, the plaintiff, Eastman Kodak Company, demands that this Honorable Court authorize the Commissioner of Patents to issue to plaintiff, Eastman Kodak Company, Letters Patent of the United States containing said claims 28 and 29 upon the filing in the United States Patent

Office of a certified copy of such adjudication, and upon compliance with the requirements of the law for such cases made and provided.

EASTMAN KODAK COMPANY,
By Clarence M. Fisher,
Attorney for Plaintiff,
National Press Building,
Washington, D. C.

N. M. Perrins,
Daniel I. Mayne,
343 State Street,
Rochester, New York.
Of Counsel.

Filed August 23, 1941.

Answer to the Complaint. 9

To the Honorable the Justices of the District Court of the United States for the District of Columbia.

I, II, III. Defendant admits the allegations of paragraphs I, II and III.

IV. He admits that by decisions rendered by the Board of Appeals of the Patent Office on May 7, 1941, and on July 1, 1941, he has refused and still refuses to issue to plaintiff a patent containing either of claims 28 or 29 of the application of Frederic R. Bean, Serial No. 134,903, which was filed in the Patent Office on April 3, 1937, and was entitled "N-Substituted Aminophenols and Motor Fuels Stabilized Therewith". He denies that plaintiff is lawfully entitled to receive a patent containing either of said claims since it is deemed that the said claims are unpatentable in view of the following prior patent and for the reasons given in the statement of the examiner's answer to the appeal and the decision of the

Board of Appeals, copies of which will be furnished at the trial:

Downing et al.,	2,009,767,	July 30, 1935,
Dahlen et al.,	2,063,151,	Dec. 8, 1936,
Atwell,	2,081,130,	May 25, 1937,
Rogers et al.,	2,084,977,	June 22, 1937,
Clifford,	2,109,164,	Feb. 22, 1938,
British patent,	350,438,	June 5, 1931.

Profert of copies of these patents is hereby made.

V. He admits the allegations of paragraph V.

W. W. COCHRAN, Solicitor,
United States Patent Office,
Washington, D. C.
Attorney for Defendant,

September 4, 1941.

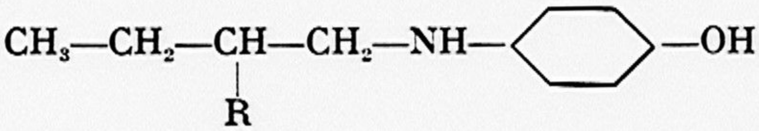
I hereby certify that a copy of this Answer to the Complaint was mailed today, September 4, 1941, to the attorney for plaintiff, Clarence M. Fisher, National Press Building, Washington, D. C.

W. W. COCHRAN,
Solicitor.

FINDINGS OF FACT 20

1. This is a civil action brought under the provisions of Section 4915 R. S. (U. S. C., title 35, sec. 63) in which it was sought to have the Court find that plainttiff, Eastman Kodak Company, as the assignee of the application of Frederic R. Bean, Serial No. 134,903, filed April 3, 1937, is entitled to have issued to it on said application a patent containing claims 28 and 29 of that application. This Civil Action was consolidated with Civil Action No. 7608, also entitled Eastman Kodak Company v. Coe, for purpose of trial, and the parties agreed that the exhibits and the testimony in the consolidated trial should serve with equal force and effect in each civil action.

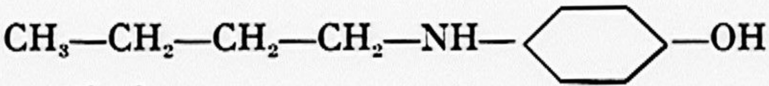
2. The application of Bean, Serial No. 134,903, involved in this action, discloses, and claims in the aforesaid claims 28 and 29, compounds having the formulas:



wherein

R is a member from the group consisting of hydrogen and an alkyl group of not more than two carbon atoms

and:



respectively.

The three compounds represented by the former formula are members of the large class known as the N-alkyl-p-aminophenols, while the specific compound represented by the latter formula is termed N-(n-primarybutyl)-p-aminophenol. Where R in the former formula is C_2H_5 the compound is designated N-(2-ethyl-n-primarybutyl)-p-aminophenol. The Bean application here in suit also discloses and claims a process for preparing such compounds; such latter claims were allowed by the Patent Office and were not in issue in this action.

3. The patent to Rogers and Voorhees, No. 2,084,977, granted June 22, 1937, upon an application filed March 26, 1937, states that the invention "relates to the use of ortho-meta-, and para-butyl aminophenols as gum inhibitors for cracked gasolines," and that "these inhibitors are aminophenol wherein one amino hydrogen has been substituted by a butyl group, particularly the normal butyl radical." The Rogers and Voorhees parent application, Serial No. 427,416, filed February 10, 1930, of which the said patent No. 2,084,977 purports to be a continuation-in-part, states that, as examples of materials to stabilize gasoline against gum formation there are employed:

> "the aminophenols, para- and ortho-methyl aminophenol, dimethyl aminophenol, hydroxyazobenzine, butyl, isopropyl and benzyl aminophenols, dihydroxy diphenylamine, paraphenetidine, amino naphthol and the like."

This quoted language of the said parent application covers N-(n-primarybutyl)-p-aminophenol. As to the disclosure of N-(n-primarybutyl)-p-aminophenol the effective filing date of patent No. 2,084,977 is February 10, 1930. Neither the said Rogers and Voorhees patent nor the said Rogers and Voorhees parent application discloses any method for preparing any of the chemical compounds recited in the foregoing quotation.

4. The British patent to Wade, No. 350,438, dated June 5, 1931, recites:

> "para- and ortho-methyl-amino-phenol, dimethyl-amino-phenol, butyl, isopropyl and benzyl amino-phenols, dihydroxy-diphenyl-amine, and the like"

as stabilizers to stabilize gasoline against gum formation. The quoted language of the said British patent covers N-(n-primarybutyl)-p-aminophenol. This patent discloses no process for preparing any of the chemical compounds recited in the foregoing quotation.

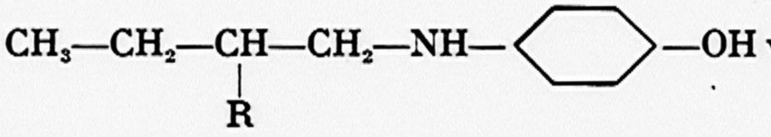
5. The patent to Clifford, No. 2,109,164, granted February 22, 1938, upon an application filed October 22, 1932, recites "N-butyl p-amino phenol," which quoted language of the said Clifford patent means N-(n-primarybutyl)-p-aminophenol, the term used in Bean's application here in suit. This Clifford patent further recites that "n-butyl p-amino phenol can be prepared by reacting butyl amine and p-chlorophenol in an autoclave." (The n in this quotation is a typographical error in the patent and should be read N.) The patent to Clifford also recites "N-isoamyl p-amino phenol" and "p-hydroxy phenyl ethyl amyl amine" (N-ethylamyl p-aminophenol). These three quoted terms are mentioned by Clifford in connection with the treatment of rubber. However, he

also refers to the stabilization of gasoline with all the substances which he names. While the plaintiff proved that the method for preparing N-(n-primarybutyl)-p-aminophenol, specified at page 3, column 2, lines 16 to 20, of the Clifford patent, No. 2,109,164, is inoperative, the plaintiff did not prove that the method referred to on page 2, column 2, lines 60 to 75, of that patent is inoperative to prepare that compound.

6. The remaining prior art relied upon by the defendant; namely, Downing et al., U. S. patent No. 2,009,767, granted July 30, 1935, upon an application filed April 28, 1933, Dahlen et al., U. S. patent No. 2,063,151, granted December 8, 1936, upon an application filed February 6, 1934, and Atwell, U. S. patent No. 2,081,130, granted May 25, 1937, upon an application filed December 30, 1932, is merely cumulative.

7. The plaintiff proved that Frederic R. Bean prepared N-(n-primarybutyl)-p-aminophenol between March 12, 1934, and September 15, 1934. The plaintiff further proved that Frederic R. Bean had prepared, under his direction, N-(2-ethyl-n-primarybutyl)-p-aminophenol on November 24, 1936. The plaintiff also further proved that Frederic R. Bean had the antioxidant efficacy of N-(n-primarybutyl)-p-aminophenol tested during the period from October 5, 1936, to October 26, 1936, and of N-(2-ethyl-n-primarybutyl)-p-aminophenol during the period from December 11, 1936, to very early 1937.

8. In view of the prior art, it was not invention to provide compounds falling within the formula:



wherein

R is a member from the group consisting of hydrogen and an alkyl group of not more than two carbon atoms.

CONCLUSIONS OF LAW

1. Claim 29 of the Bean application, Serial No. 134,903, is anticipated by the British patent to Wade, No. 350,438, the patent to Clifford, No. 2,109,164, and the patent to Rogers and Voorhees, No. 2,084,977, and claim 28 thereof is not patentable in view of this prior art.

2. Plaintiff is not entitled to a patent containing either of the claims 28 and 29 of the said Bean application.

3. The N-(n-primarybutyl)-p-aminophenol of Bean claimed in claim 29 stands anticipated by the British patent to Wade, the patent to Clifford, and the patent to Rogers and Voorhees, regardless of whether this prior art does or does not disclose a process for preparing the compounds, and regardless of whether or not this prior art discloses a process of preparing these compounds which is operative.

4. Bean having proved reduction to practice of N-(n-primarybutyl)-p-aminophenol and of N-(2-ethyl-n-primarybutyl)-p-aminophenol prior to March 26, 1937, the filing date of the Rogers and Voorhees patent, No. 2,084,977, that patent is prior art against Bean, only insofar as the Rogers and Voorhees parent application, Serial No. 427,416, filed February 10, 1930, discloses N-(n-primarybutyl)-p-aminophenol.

5. The complaint should be dismissed.

D. W. O'DONOGHUE,
Justice.

25

Final Judgment.

This action came on to be heard at this term; and thereupon upon consideration thereof, it is this 17th day of November, 1941, adjudged that the complaint be, and it is hereby dismissed with costs against the plaintiff.

D. W. O'DONOGHUE,
Justice.

Approved as to Form:
C. M. Fisher,
Attorney for Plaintiff.

Evidence on Behalf of the Plaintiff

By Mr. Mayne: I would like to offer in evidence as Plaintiff's Exhibit 1 a certified copy of Bean Application 134,903, * *. 34

(Certified copy of Bean Application 134,903 was marked Plaintiff's Exhibit No. 1 and received in evidence.)

DR. FREDERIC R. BEAN was called as a witness on behalf of the plaintiff and, being first duly sworn, was examined and testified as follows: 39

DIRECT EXAMINATION By Mr. Mayne:

Q. Will you state your name, address, and occupation? A. My name is Frederic R. Bean. My occupation is chemist, and I live at 109 Heberton Road, Rochester, New York.

Q. Are you employed by anyone at the present time? A. By the Eastman Kodak Company.

Q. How long have you been a chemist, Doctor Bean? A. I went with the Eastman Kodak Company in 1931 and I have been with them ever since.

Q. Just state briefly the nature of your academic training before you went there. A. I took my undergraduate work at DePauw University, Green Castle, Indiana, and majored in chemistry, and then took my graduate work at Cornell University and majored in organic chemistry.

Q. From the time you were employed by Eastman Kodak Company, commencing in 1931, what has been the nature of your duties there as a chemist? A. They have to do with research and development, the study of photographic developers in the aminophenols and all other chemicals used in the photographic industry.

40 Q. Are you the same Frederic R. Bean who is designated as applicant in Plaintiff's Exhibits 1 and 2? A. Yes; I am.

* * * *

42 Q. Will you please describe to the Court, just briefly, what your notes record there or, in fact, what you recall from the preparation you made about March 12, 1934? A. The preparation itself you want described?

Q. Just describe what happened there. A. Well, I was interested in getting this particular type of compound of alkyl para aminophenol, and I started out with acetaldehyde and carried on through to butyraldehyde. In the case of the butyraldehyde I dissolved para aminophenol in a caustic alkali solution and added butyraldehyde to that and let it cool to an ice cold temperature and then added activated aluminum to that, and that produced this nitrogen normal primary butyl para aminophenol as shown by the melting point and the general method of synthesis.

Q. In this reaction you have just described you combined the butyraldehyde with para aminophenol, you said? A. Yes.

Q. Would it be possible, if that reaction goes to completion, for another product to occur than the nitrogen normal primary butyl para aminophenol? A. Not in any quantity. About the only one I can postulate and the only one I can get is the butyl para aminophenol. I cannot get anything else.

Q. The use of activated aluminum: was that the usual procedure, or did that involve some difficulties? A. The making of activated aluminum is a very complicated procedure and requires a considerable bit of experience and practice to handle. If it is not properly handled, reduction will not proceed at all, and if it is over-activated it will not proceed. You have to get the proper state of activation of the aluminum.

* * * *

Q. And what were the results of the nitroso reaction you conducted at this time? A. Well, there was an immediate precipitation of the nitroso derivative of nitrogen-(normal-primary butyl)-p-aminophenol. 46

Q. And did that serve as proof to you with respect to this compound here that we have been talking about? A. Yes. It definitely proves that that is a secondary amine and the relation to the synthesis—method used for synthesis—proves that that is the compound that I sought, the nitrogen-(normal-primary butyl)-p-aminophenol.

* * * *

Q. Now, is there anything else in this notebook V of yours which has any relation to this subject you have been testifying about? A. Well, I rechecked the melting points of the normal-primary butyl-para-aminophenol base and rechecked the nitroso derivatives, and then I also tried the solubility to determine whether or not there were sufficient solubility to butyl compounds, sufficient solubility to be incorporated in gasoline. 50

Q. On what page do you find the record with respect to gasoline solubility? A. On page 72.

Q. Will you tell the Court, please, just what that records with respect to nitrogen-(normal-primary butyl)-para-aminophenol? A. It records that five-thousandths of a gram of the nitrogen-(normal-primary butyl)-para-aminophenol dissolved in 50 cubic centimeters of gasoline.

Q. Did you regard that as very spare solubility or exceedingly soluble, or how can you characterize that? A. I remember and also recalled that it is quite soluble in the gasoline;

I mean, sufficient solubility for the purpose which I sought.

Q. Why were you interested at that time in determining the solubility in gasoline of this compound? A. Well, I was interested in it as a gum inhibitor or an antioxidant to be used to stabilize and preserve gasoline.

* * * *

54 Mr. Mayne: I might state, your Honor, at this point, that the Solicitor has been good enough, on several letters, to agree to the authenticity of them and of the transmittal of those letters and receipt by the addressee. On some of those formal matters we would have had to have brought probably half a dozen additional witnesses; and he does not admit, of course, the actual chemical identity of the samples that are mentioned in some of these letters, but as to the actual transmittal of the letter and receipt of it he has agreed to stipulate that those letters were transmitted and received. This is one of those, in that he agrees, I understand, that the letter was received by Mr. Dickins, the addressee, and I offer that in evidence as Plaintiff's Exhibit No. 6.

(Typewritten letter dated May 10, 1935, from F. R. Bean to A. W. M. Dickins, with attached photographs, was marked Plaintiff's Exhibit No. 6 for identification.)

* * * *

57 Mr. Mayne: I offer this letter in evidence as Plaintiff's Exhibit No. 7, this letter being dated October 5, 1936.

(Longhand letter dated October 5, 1936, from F. R. Bean to D. I. Mayne, was marked Plaintiff's Exhibit 7 and received in evidence.)

Mr. Mayne: There are two further letters, your Honor, which come under our stipulation. One is a let-

ter of October 2, 1936, from D. I. Mayne to James C. White, Tennessee Eastman Corporation, discussing this subject. The other is a letter dated October 6, 1936, sent 58
in the name of D. I. Mayne but initialed by "BW," addressed to James C. White, Tennessee Eastman Corporation, transmitting three samples, of which one is nitrogen-butyl-para-aminophenol sulphate. These two come under our stipulation as having been transmitted.

Mr. Moore: Yes.

Mr. Mayne: I offer these in evidence: the letter of October 2 as Exhibit No. 8.

(Typewritten letter, October 2, 1936, from D. I. Mayne to James C. White, was marked Plaintiff's Exhibit 8 and received in evidence.)

Mr. Mayne: And the letter of October 6 as Exhibit No. 9.

(Typewritten letter, October 6, 1936, from D. I. Mayne to James C. White, was marked Plaintiff's Exhibit 9 and received in evidence.)

By Mr. Mayne:

Q. I hand you a form, Doctor, entitled "Sketch Sheet" and bearing a number in the upper right-hand corner of 24266. Can you tell us anything about that? A. This is a form which we generally use for filling out or making an application for —you might say, drawing up a draft from which an application can be made for a patent.

Q. What does this sketch sheet No. 24266 describe, Doctor? A. Well, in this sketch sheet I have described the preparation of N-butyl-para-aminophenol and the use thereof as an antioxidant inhibitor, and I give the melting points of the base and of the nitroso derivative, and claim its being useful as an antioxidant for preventing the deterioration of oils and fats 59 and fatty oils and fatty acids and fatty esters and in the stabilization of gasoline, petroleum distillates.

Q. What date do you find on this sketch sheet? A. The sketch sheet was drawn up May 27, 1935, and the witness is August 20, 1936.

Q. Is that your signature on the lower right-hand corner? A. Yes.

Q. Whose signatures appear as witnesses to this sketch sheet? Can you tell me? A. Austin J. Gould; he worked in one of the laboratories closely associated with mine. And Dr. Leo Tanghe; his signature is there.

Q. Now, on the back of this sketch sheet what do you find there? For instance, what do we mean by your entry there after "Earliest date of invention"? A. Well, that was the earliest date at which I had made this compound, this nitrogen-butyl-para-aminophenol, March 12, 1934.

* * * *

60 Mr. Mayne: I will offer this sketch sheet No. 24266 in evidence as Plaintiff's Exhibit No. 10.

Mr. Moore: Same objection as to Plaintiff's Exhibits 2, 3, and 4.

The Court: I will receive it in evidence for whatever probative force it may have.

(Sketch sheet dated May 27, 1935, was marked Plaintiff's Exhibit 10 and received in evidence.)

By Mr. Mayne:

Q. Did you ever get a report on the testing of the antioxidant properties of this nitrogen-normal-butyl-para-aminophenol? A. Yes.

Q. I hand you a letter dated December 15, 1936, purportedly from D. I. Mayne to A. W. M. Dickins, and ask if you ever saw that letter or a copy of it before. A. Yes; that is the same letter I have. I have a copy of it. 61

Q. Did you receive a copy of that letter currently with the date thereof? A. I received a copy of it, yes.

Q. What does this appear to report? A. Well, it shows definitely that nitrogen-butyl-para-aminophenol is an excellent antioxidant to be used in gasoline.

* * * *

Mr. Mayne: Also under our stipulation I offer in evidence as Plaintiff's Combined Exhibit No. 12 a letter from James C. White of the Tennessee Eastman Corporation, dated October 26, 1936, to D. I. Mayne at Rochester, N. Y., this letter having attached thereto a report numbered 602-6, entitled "Analysis of TE-2 and TE-4", which I have shown to the Solicitor and am giving photostatic copies, that being the report that is dealt with in the letter which is dated December 15, 1936, from D. I. Mayne to A. W. M. Dickins. 62

(Typewritten letter dated October 26, 1936, from James C. White to D. I. Mayne, and attached report No. 602-6, were marked Plaintiff's Exhibit No. 12 and received in evidence. 63

By Mr. Mayne:

Q. Now, this letter of December 11, 1936, from Dickins to Perrins, about which you were testifying: I notice it speaks of a sample of 2-ethyl-butyl-p-aminophenol (base). How does that figure in your work at that time, if at all? A. Well, we were interested in going on with production of these aminophenol derivatives of that type and in introducing a larger group on the nitrogen, and in general in determining the behavior of such a compound and the effect the introduction of that group would have on the compound: the solubility, whether its potency was changed, and so forth.

Q. Did you prepare or have prepared under your direction at any time any of this 2-ethyl-butyl-p-aminophenol? A. Yes.

Q. Just tell us about the circumstances, will you, please? A. Well, I had an assistant to continue this work on the reduction of this type of compound, the aldehyde aminophenol with activated aluminum, and had one of my assistants prepare some of this compound, 2-ethyl-normal-butyl-aminophenol.

Q. Who prepared that compound for you? A. Mr. Vaughn.

Q. And about what time did he prepare that compound with relation to this letter, for instance? A. Well, sometime preceding it; I don't know specifically the date there.

64 Q. This is dated December 11, 1936? A. Well, he prepared it previous to that.

* * * *

Q. After this sample of ethyl-butyl-aminophenol went forward
65 through the company channels to Tennessee Eastman Corporation, did you get any evidence that they had tested the antioxidant properties of this compound as requested? A. Yes.

Q. How did that evidence come to you, Doctor? Do you remember? A. Well, it was brought in the form of the application for the patent and placed in that, and that is the way I had received it at that time.

Q. That information on the antioxidant properties, then, was incorporated in the application, was it? A. Yes.

Q. And was in the application at the time you signed it? A. Yes, sir.

* * * *

70 Q. You have testified as to several identification tests or means by which you know that these compounds that you prepared or had prepared are the ones in question. What other identification tests have been made by you or under

your direction, either currently with the preparation of the samples or since? A. We have made, or I have, an analysis of this compound.

Q. What was the nature of those tests? That is, were they some more nitroso reactions, or just what were they? A. It was the determination of the nitrogen, carbon, and hydrogen 71
in the nitrogen-normal-primary-butyl-para-aminophenol.

Q. Who carried out those tests? A. I didn't understand.

Q. Who carried out or supervised those tests? Can you say? A. My assistant, Mr. Vaughn.

Q. He is here in the court room, is he? A. Yes.

Q. Are you familiar with a U. S. patent to one Clifford, No. 2,109,164, which is one of the patents cited in the defendant's answer? A. May I see the patent?

(The patent referred to was handed to the witness.)

The Witness. Yes.

By Mr. Mayne:

Q. At any place in that patent does it use the name "butylaminophenol"? A. Yes.

Q. Where in that patent does he use the name "butyl aminophenol"? A. I find on page 3, the upper part of the second column, the mention of this compound.

Mr. Moore: You haven't numbered that.

Mr. Mayne: That, your Honor, is item No. D in Defendant's Exhibit No. 1.

Mr. Wiczer. No. E.

Mr. Mayne: Would you indicate the lineage? Tell us 72
what line and page that is.

The Witness. Well, it is between 15 and 20 on page 3.

By Mr. Mayne:

Q. Which column? A. On the right-hand column.

Q. What does he say there about butyl aminophenol? A. He says, "Thus n-butyl p-amino phenol can be prepared by reacting butyl amine and p-chlorophenol in an autoclave."

Q. Do you find anything else in this patent whatsoever that refers to the preparation of this compound? A. No.

Q. Does he say anything whatsoever in this patent, other than what you have read, as to the preparation of any butyl aminophenol? A. No.

Q. You testified, I believe, that you had been working with this general character of chemistry since about 1931, didn't you? A. Yes.

Q. What can you say about that reaction, namely, butyl amine with para-chloro phenol in an autoclave, as to its general application? A. Well, in the description there there is a statement, of course, that it was prepared by reaction of butyl amine para-chlorophenol in an autoclave, and we usually—

Q. (Interposing) Well, what I am getting at is: Take the known compound; that is, the one that had been known, I understand, for a long time before your work, the methyl 73 aminophenol. Was that reaction which is referred to on page 3 by Clifford a known type of reaction for the methyl aminophenol? A. Yes, it had been used with methyl aminophenol.

Q. And might a man, or a chemist—assuming a chemist knowing that that reaction had been used to prepare methyl aminophenol, might he suggest that for butyl? A. Yes. The idea would be, in general, to try what to me were conditions favorable for production of methyl aminophenol on that reaction, from my experience with that type of reaction.

Q. Did you do anything to test out the validity or accuracy, let us say, of what Clifford has suggested on page 3 here as a method of preparation. A. Yes.

Q. When did you carry that work out or have it carried out? A. Well, as I recall, it was 1938 we started on the work with the butyl amine and the chloro phenol.

Q. In general was that work carried out by you or by one of your assistants? A. My assistant did the greater part of the work. I ran some runs of my own and then turned it over to him to check further.

Q. I expect to have him testify on the details, but would you just tell us generally what it was you did or had done there to test this reaction out? A. Well, I placed a butyl amine in equimolecular proportions with para-chloro phenol in an autoclave and heated it at various temperatures from a hundred up to well above two hundred degrees under those 74
various conditions; and we varied, I pointed out there, the temperature over quite a range, to give it a good chance or a fair trial of trying to make the thing proceed as nearly as we could deduce from this statement in the patent.

Q. And were you able to get a reaction? A. We obtained absolutely no butyl para-aminophenol out of that reaction, or out of that autoclave, I mean.

Q. Now, if I may be permitted just to summarize there at one point, you put your butyl amine and your para-chloro phenol together in the autoclave as full-strength compounds? A. Yes.

Q. At, say, 200, was it? A. Yes.

Q. Centigrade? A. Yes.

Q. And tried to get a reaction? A. Yes.

Q. Now, from that, as one interpretation, what did you do to give it a further chance to react? What variation did you employ next, for instance? A. Well, I placed a copper catalyst, copper sulphate as a catalyst, in the reaction mixture in the hope that that would help.

Q. What made you think of the copper catalyst idea? A. Well, that has been used in that type of reaction in the replacing of a halogen with amines, or ammonia, for that matter, on the aromatic nucleus.

Q. Does Clifford say anything about a copper catalyst? 75
A. No. And further than that, we extended it to placing

water in there in the hope that that also would create a more favorable condition.

Q. Reactions sometimes go better in aqueous media, do they? A. Yes. In that type of reaction frequently, if it is anhydrous, which is the impression you get from his statement there—the impression I get from his statement here—if you heat them up you get a tar formation; and, well, it is usually just a very indefinite tarry matter that you obtain out of such a reaction.

Q. And you applied to the reaction mixture, after it had been given an opportunity to react, a test to determine whether there was any aminophenol? A. Yes, we went very thoroughly into determining whether or not there had been any butyl paraaminophenol, determined by all the tests that we knew.

Q. Now, that was, you say, with butyl amine. Was that a normal butyl, or was it an isobutyl, or what butyl? A. It was the normal butyl amine, primary butyl amine.

Q. And did you try it out with any other forms of butyl amine? A. We tried it with isobutyl amine.

Q. And did you get any favorable results there? A. We got none whatever.

Q. Then may I put it this way, Doctor: You have used every reasonable effort, have you, to make this Clifford reaction go?
76 A. I feel that I have thoroughly tested the thing, in the light of my knowledge of that type of reaction, given it every chance possible, and tried every way I could to make it work.

Q. I hand you a two-page report dated October 10, 1941, entitled "Report on the Investigations of the Clifford U. S. Patent 2,109,164," and ask if your signature appears on that report. A. Yes.

Q. Do you confirm all that is in that report? A. Yes, sir.

Mr. Mayne: I should like to mark this for purposes of identification as Plaintiff's Exhibit No. 15.

(Report on Investigations of Clifford Patent was marked Plaintiff's Exhibit 15 for identification.)

* * * *

Cross Examination by Mr. Moore:

Q. You knew at the time that you made these tests and made this report, that is, Plaintiff's Exhibit— A. (Interposing) Which test are you referring to? 78

Q. The tests involved in Plaintiff's Exhibit No. 15 that were the basis of that report. A. The Clifford?

Q. Yes. You knew at that time that Clifford had been cited as a reference against your application, didn't you? A. Yes.

Q. Now, you have read this Clifford patent with care? A. Well, yes, I have looked through it and read it.

Q. Now, you refer to a process that is set forth on page 3, the second column, beginning line 16. A. Yes.

Q. That is a general or a type reaction that is set forth there, isn't it? A. Yes.

Q. And you would expect to use the skill and knowledge and artistry that you acquired by your training and experience in following out that process? A. Yes, sir.

Q. Now, you say that it was old to use methyl or an ethyl amine in such a process? A. Yes.

Q. As starting material? A. Yes.

Q. Did you test your experiments by using ethyl or methyl amine? A. I have done numerous experiments with methyl amine reaction. 79

Q. Did you at this time? A. Precisely at this time?

Q. Yes. A. Well, a time preceding that. That is where I got a great deal of my experience in that type of reaction.

Q. Will you refer to the Clifford patent, page 2, second column, beginning line 59? A. Yes.

Q. Doesn't Clifford there give another method of preparing the antioxidants that are the basis of his patent? A. Not in my experience can this information he gives here on your citation there, 59, be applied in any way whatever to the use of alkyl amines.

Q. He does give that information—he does refer to an alternative method at that point? A. Not for alkyl amines.

Q. He describes a preparation of para-hydroxy diphenyl amines there, does he not? A. Yes.

Q. And this method involves reaction of analine in hydroquinone? A. Yes.

Q. And analine is an aromatic amine, isn't it? A. Yes.

80 Q. And as a chemist wouldn't you expect to use the reaction there stated to prepare other aminophenols, such as butyl aminophenol? A. No, sir.

Q. I hand you the Von Bramer patent, 2,163,639, which is assigned to the Eastman Kodak Company, and ask you if that doesn't actually disclose your butyl aminophenol you prepared by reacting the butyl aminophenol with hydroquinone.

> Mr. Mayne: May it please the Court, there has been no notice on this; it hasn't been set up in the answer. I really have no objection to his being examined, except that we have had no chance to look over that subject.
>
> Mr. Moore: I just do it to support the validity of the Clifford patent which has been attacked by testimony here, your Honor.
>
> The Court: I think he may properly cross-examine and call his attention to another patent issued to the plaintiff that involves some similar operation to the patent involved in this case.

The Witness: Will you point out to me the case, the instance?

Mr. Wiczer. Example 1.

Mr. Moore. Example 1.

The Witness. Will you repeat?

By Mr. Moore:

Q. Now, that discloses that your butyl aminophenols are prepared by reacting the butyl amine with hydroquinone? A. Yes.

Q. Is that right? A. Yes, sir.

Q. Isn't this a method you could follow to prepare your butyl aminophenol? A. Yes, you could prepare my butyl phenol that way, but it would not be as pure by any means; you get certain by-products in there. 81

Q. Then couldn't you in a sense use one method of the Clifford patent to prepare your butyl aminophenol?

Mr. Mayne: Well, but, your Honor, this patent was filed in 1936. It is incompetent. I do not object to examination, but it is entirely incompetent, because Clifford was filed in 1932 and we are speaking as of the date of the Clifford patent and not of something that happened since.

Mr. Moore: We are also speaking as of a time prior to the filing of the Bean application, April 3, 1937.

Mr. Mayne: Well, I submit it is not competent.

The Court: You may cross-examine on the way different chemicals may be used, put together.

By Mr. Moore:

Q. Isn't the method described in the Von Bramer patent which I showed you a specific example of the general method set forth on page 2 of the Clifford patent? A. You say that the two, you think—you ask me—

Q. I ask you, in your opinion aren't they two related to the Von Bramer process, being a specific example of the general method set forth in Clifford? A. No; the analine will not work at all under the conditions which this primary butyl amine will. Hydroquinone—

Q. (Interposing) But the analine is an aromatic amine? A. It is an aromatic amine, and butyl amine is an aliphatic amine, so there is an extreme difference there. 82

Mr. Moore: That is all.

By the Court:

Q. In this Clifford patent on page 2, specifications line 8, isn't there a mention there of N-butyl p-amino phenol? A. Clifford? Page 2?

Q. Clifford, page 2, line 8. A. Second column. There is mention of nitrogen-butyl para-phenol. That doesn't tell me what that compound is at all, from my knowledge of organic chemistry. I have no knowledge of what kind of a butyl group that was, what its properties were, or anything else.

Q. Mr. Merchant mentions that line there in his opinion, Mr. Bean. Weren't you aware of the fact that the Examiner in his decision in your case mentioned that line and that paragraph there? You have stated that the only time it was mentioned was on page 3, second column. A. No.

Q. The 18th line. A. No, your Honor. I think I mentioned there that the preparation is mentioned in only one place.

Mr. Moore: May I make a statement, your Honor? There is a mistake in that page three. The small n should be large N. That is merely a printer's error. Everyone accepted that.

The Court: The same thing?

Mr. Moore: Yes.

The Court: It was mentioned on page 2?

Mr. Moore: Yes, your Honor.

83 By the Court:

Q. Now, there have been a number of exhibits offered here by the plaintiff, being letters either written by or to Mr. J. C. White and, Mr. D. I. Mayne, Mr. N. M. Perrins, Mr. A. W. M. Dickins, and by you. All of these parties mentioned, including yourself, are employees or officers of the Eastman

Kodak Company, are they not? A. Yes, sir.

Q. And on salaries received from the plaintiff in this case, the Eastman Kodak Company? A. Yes, sir.

Q. You have written letters among yourselves from one to the other with regard to your patents involved in this case? A. Yes. In the usual manner, yes.

The Court: That is all.

By Mr. Moore:

Q. And in these letters you have used, have you not, the same description of the compound that you say you produced back in March 12, 1934, as that description used by Clifford, used in his patent? Let me show you your notebook (indicating). A. Yes.

Q. Exhibit 5. A. Yes.

Q. A letter to Mr. Dickins, signed by you, referred to the title and page 1 and numerous places on page 2? A. Yes. I can explain that.

Q. You have used the same language as Clifford? A. I have used, to me, what you would call a shortened terminology, and my use of that is based on my preparation. I know what I've got, and I have the melting points of it, and it is a very definite thing to me, and it is no more than if I sent it down to them marked with "X". I was just interested in the results. 84

The Court: The question was whether your description in those papers was the same as in the Clifford one. That was your question?

Mr. Moore: That is the question, your Honor.

By the Court:

Q. Will you answer that question? A. Yes.

Q. It is the same? A. But the meaning back of it, to me, is: To me, I prepared this compound on the basis of synthesis, and

I know the structure of it, and I refer to that, and I can't see where the name has any bearing on it.

By Mr. Moore:

Q. You used that same language in a letter to Mr. Mayne? A. Yes.

Q. Signed by you? A. Yes.

Q. And Mr. Mayne is a member of the Patent Department of the Eastman Kodak Company? A. Yes.

Q. And the man who examined your—

By the Court:

Q. (Interposing) What is the date of it? A. That is October 15, 1936.

85 The Court: What exhibit?

Mr. Moore: Exhibit 7, your Honor.

The Witness: Seven.

By Mr. Moore:

Q. In Exhibit 10, the sketch sheet, your conception data sheet, you used the same language, didn't you? A. Yes, we used that language.

The Court: By "the same language" you mean the language used in Clifford?

Mr. Moore: The language used by Clifford, yes.

The Court: In his specification?

Mr. Moore: The language used by Clifford in his specification on page 2, column 2, line 8.

The Witness: Well, it's a case of meaning differently to me than it does to—

By the Court:

Q. (Interposing) The language is the same as in Clifford? A. Yes, that is true. It is printed that way.

The Court: Any further cross-examination?

Mr. Moore: No further cross-examination.

Re-direct examination by Mr. Mayne:

Q. Just one point. Back of this name business you had a method of preparation, did you not, Doctor? A. Yes, sir.

Q. And that method of preparation was described in connection with your mention of nitrogen-butyl-para-aminophenol in a number of cases, was it? A. Yes, sir.

Q. And would that, to you, or to anyone skilled in the art, 86
also knowing the method, mean a broad compound, or would that mean the compound that resulted from the synthesis? A. It would mean the compound from the synthesis.

* * * *

DR. LEO J. TANGHE was called as a witness on behalf of 87
the plaintiff and, being first duly sworn, was examined and testified as follows:

Direct Examination by Mr. Mayne:

Q. Where do you live? A. 120 Barnard Street, Rochester, New York.

Q. What is your present occupation? A. I am a chemist at the Eastman Kodak Company.

Q. How long have you been so employed by the Eastman Kodak Company? A. I have been employed as a chemist
there since 1930 continuously, with the exception of some time 88
taken out for further education.

Q. Prior to 1934 what had been the extent of your chemical education, Doctor? A. I had taken my Bachelor's and Master's degree in chemistry at the University of Rochester.

Q. Up to the fore part, or let us say prior to 1934, what if anything had you had to do with the preparation of amino compounds or anything related to that? A. When I first came to the Eastman Kodak Company I worked on photographic developers and amino compounds for a few months.

Q. Are you acquainted with Dr. Frederic Bean who has just been testifying? A. Yes; I am.

Q. When did you first become acquainted with Dr. Bean? A. When he first came into the Eastman Kodak Company, which, I believe, was in 1931. I have known him since then.

Q. Did you ever work in the same room or laboratory with Dr. Bean? A. Yes. I worked in the same laboratory with him for the major portion of the time from 1931 to 1934.

Q. Were you connected with the Eastman Kodak Company and working in the same laboratory with Dr. Bean in 1934? A. Not for the entire year of 1934. I was, until about the middle of September, working with Dr. Bean in the same laboratory.

Q. What happened in the middle of September, 1934? A. In the middle of September, 1934, I left the company temporarily to continue education at the Ohio State University. 89

Q. Was that further education in chemistry? A. Yes; it was.

Q. Did you subsequently receive your Doctor's degree in chemistry? A. Yes, I did, in 1937.

Q. Did you during 1934 and prior to your departure from there on September 15, 1934, see Dr. Bean prepare a compound known as nitrogen normal primary butyl para aminophenol? A. Yes. I saw him make preparations of that compound in the laboratory.

Q. Can you tell us what process he employed in order to prepare that compound? A. He employed the process of reacting para aminophenol with an aldehyde in the first step of the reaction and then reducing that intermediate compound with aluminum to obtain the primary normal butyl para aminophenol.

* * * *

91 Mr. Moore: No cross-examination.

By the Court:

Q. Just what, in your opinion, was the discovery that was made by Dr. Bean in getting this result which is in Exhibit 5? A. In my opinion, he was the first person to have this butyl para aminophenol in question.

Q. How was it that it was something new that was never 92
known before? What did he do that nobody else had ever done before? A. He put these various chemicals together under favorable conditions for the formation of the compound and isolated it as a fairly pure substance.

Q. What chemical did he put in that no one else had ever put in? A. He used a method involving para aminophenol with butyraldehyde. Previous mentions of the compound are not very definite as to what was placed in the reaction to obtain the compound or whether it had been obtained, even. I think his work shows conclusively that he actually had this compound and that point can hardly be questioned from the way in which the synthesis was carried out.

Q. Did not the prior art suggest that he might have obtained this resultant? A. By carrying out the reaction as Dr. Bean has?

Q. Yes; or like Clifford suggested, or like any other expert chemist might suggest. A. No; I do not think the prior art did.

Q. Can you tell the court just what he did that nobody else had done or suggested before? A. He actually carried out the synthesis on this compound in a way which, so far as I know, had not been suggested before.

Q. What was that way? A. By condensing an aldehyde with para aminophenol and reducing the compound—reducing the intermediate compound.

The Court: That is all. 93

STODDARD VAUGHN was called as a witness on behalf of the plaintiff and, being first duly sworn, was examined and testified as follows:

Direct Examination by Mr. Mayne:

Q. Where do you reside, Mr. Vaughn? A. 82 Merchant Road, Rochester, New York.

Q. What is you occupation? A. Chemist.

Q. Are you employed by anyone? A. The Eastman Kodak Company.

Q. How long have you been so employed? A. Since February 1, 1936.

Q. What academic training had you before you went with the Eastman Kodak Company? A. I received my Bachelor of Science degree from the University of Maine, in chemistry.

Q. When you joined the Eastman Kodak Company what was the first chemical work that you were assigned to there? A. The very first compounds that I worked with were to be used as fur-dyes, and they are also closely related; that is, the compounds that are fur-dyes are also photographic developers and antioxidants.

Q. For whom did you work when you first went to the Eastman Kodak Company? A. I was directly under Dr. Bean.

94 Q. Do you still work with Dr. Bean? A. Yes.

Q. In so far as these two actions are concerned, what materials did Dr. Bean seem to be interested in at the time you went there in 1936? A. From the first month that I worked there it was with fur-dyes, and then I switched to gasoline antioxidants.

Q. Was he just doing a little work on that, or a lot of work, most of his time, or what? A. Well, naturally our work is in several fields. I would say that at that time he was devoting possibly a third of his work to gasoline antioxidants.

Q. At any time, let us say prior to 1937, did you prepare any compound of the aminophenol type for Dr. Bean? A. I prepared a great many of them.

Q. Did you prepare any aminophenol that had any sort of butyl radical in it? A. I prepared nitrogen-2-ethyl butyl para aminophenol.

* * * *

Q. After you got the sulphate what did you do, according to this record, if anything? A. I made the free base of nitrogen-2-ethyl-butyl-para-aminophenol. 96

Q. Do I understand that you converted the sulphate of all the compound you prepared to the free base, or part of it, or what? A. I converted all of it.

Q. Did you apply any test to that compound to determine the identity of the compound at that time? A. The melting point in this reaction would be unusually significant, in that if the reaction had not proceeded satisfactorily the para aminophenol that was one of the starting ingredients would have been the product we obtained rather than the combination of the aldehyde and para aminophenol, and the melting point of the para aminophenol is about 186 degrees; so that when the melting point was taken and it was about 98 degrees, I believe, it certainly showed that the desired compound had been obtained.

* * * *

Q. Mr. Vaughn, have you ever conducted or directed the conduct of any analysis of the compound of Exhibit No. 3 or the compound of Exhibit No. 17, which is that small vial? A. Yes; I have. 99

Q. What was the nature of that, taking Plaintiff's Exhibit No. 3 to be orderly? A. The free base or the sample in that bottle and the sample contained in Exhibit No. 17 was analyzed for carbon, hydrogen, and nitrogen content.

Q. When did you do that work or have it done? A. I think it was the third of October, 1941.

Q. Did you make that analysis and sign a report on that subject? A. I did.

Q. I hand you report dated October 10, 1941. Is that the report to which you refer? A. Yes.

Q. What did you find, Mr. Vaughn, as to the carbon, hydrogen, and nitrogen content of the free base which you state you took from Exhibit 3? A. I do not recall the exact percentages. 100
May I see the report?

(A document was handed to the witness.)

A. (Continuing) The nitrogen normal primary butyl para aminophenol contained 72.73 percent carbon against a calculated or theoretical content of 72.7, which is exceedingly close. The hydrogen found by analysis was 9.07. The hydrogen as calculated was 9.15. The nitrogen found by analysis was 8.58 and checked as 8.53; calculated as 8.48, which, in my mind, leaves no doubt that the compound in question is nitrogen normal butyl para aminophenol.

Q. How did this compound turn out with respect to some of the Exhibit 17 compound, the 2-ethyl-butyl? A. The percentage of carbon as calculated was 74.6; found by analysis 74.68. The percent of hydrogen calculated was 9.92. Found by analysis 9.99. The percent of nitrogen calculated was 7.25; found 7.42, and checked as 7.45.

* * * *

101 Q. Are you familiar with the Clifford patent, U. S. No. 2,109,164? A. I am.

Q. Will you turn to the right hand column on page 3, commencing at line 18? Have you ever repeated the reaction that is described in that line and the succeeding few lines? A. Line 17?

Q. Commencing at line 17. A. Yes; I have.

Q. Did you collaborate in any way with Dr. Bean on that? A. I did.

Q. I hand you report marked "Plaintiff's Exhibit 15", dated October 10, 1941. Does your signature appear on that report?
102 A. Yes.

Q. Is that a correct description of the work which you carried out or collaborated with Dr. Bean on? A. It is.

Q. The method described there with respect to the primary butyl compound—was that just a simple autoclaving of butyl amine with para aminophenol? A. That was the first reaction tried, but we tried several others. In the Clifford patent it is merely stated that the two are put together in an autoclave. No temperature is specified. The use of a catalyst is not mentioned. The use of an aqueous medium is not mentioned. But we repeated at various temperatures running from 100 to 270 degrees Centigrade. We repeated using aqueous medium and also using copper powder and copper sulphate and mixtures of both.

Q. With any of the methods or variations that you testified you tried, were you successful in obtaining any butyl aminophenol of any kind? A. No, sir.

Q. How did you check that answer, by what test, if any? A. Well, first we tried to isolate it, first with sulphate or free base. We were unable to do it, so we tested the liquors for the smaller quantities that might have formed by a nitroso test, but all results by this test were negative.

Q. From all your experience to date would you be able to suggest anything to make the Clifford process work? A. To make it work?

Q. Yes; as he has defined it in the second column on page 3. A. You mean, using certain refinements, or just with the information that is given here? 103

Q. Yes. A. No.

Mr. Mayne: I offer in evidence the exhibit previously marked as Plaintiff's Exhibit 15.

(A report dated October 10, 1941, previously marked Plaintiff's Exhibit No. 15 for identification, was received in evidence.)

By Mr. Mayne:

Q. Will you tell us a little bit more about your ideas on this nitroso reaction? Have you tried that out with anything other than just the nitrogen normal primary butyl para-aminophenol? A. Oh, yes. We tried it out on every secondary amine we prepared, and we have prepared a good many of them.

Q. Did you ever try it out on the isobutyl compound? A. I have.

Q. Is it an indicator of aminophenol formation? A. Yes. A very definite precipitate forms even when minor quantities are used. And in relation to the work we did with the Clifford patent we even went so far as—we thought it might be a possibility that the presence of small quantities of unreacted p-chlorophenol might interfere with the test, so we went so far as to make up small quantities containing both p-chlorophenol and nitrogen primary normal butyl-p-aminophenol so that we could be sure there was no interference with it.

Mr. Mayne: That is all.

104 Cross-Examination by Mr. Moore:

* * * *

Q. Referring to Exhibit 16 your notebook, with the instructions that Dr. Bean had given to you about the process of preparing butyl para aminophenols you had no difficulty in preparing that 2-ethyl-butyl-para-aminophenol described on that page, did you? A. No.

Q. It was just a routine matter? A. Well, except the activation of the aluminum.

Q. That requires the skill of a chemist? A. A little more than that. Any chemist could not go into the laboratory and 105 activate that aluminum properly. I mean, he could not guarantee that he could.

Q. Where did you receive your instructions as to that? A. I practiced it.

Q. After Dr. Bean had told you to do it, or had you had previous experience? A. No; I had never had previous experience. I practiced before I tried it on a run.

Q. You knew that the aluminum had to be activated from the instructions received from Dr. Bean, and to prepare yourself for the test you did practice activating aluminum? A. That is right.

Q. And that required a skill which perhaps the ordinary chemist would not have at first, but which he could obtain? A. It is nothing that he could not acquire.

Q. Referring to Plaintiff's Exhibit 19, did the test that you ran in the preparation of that report give you any idea as to whether or not the para butyl aminophenol prepared would be a normal or an iso form? A. No, sir.

Mr. Moore: That is all.

DR. LEE G. DAVY was called as a witness on behalf of the plaintiff and, being first duly sworn, was examined and testified as follows:

Direct Examination by Mr. Mayne:

Q. Where do you reside, Dr. Davy? A. At 1604 Pinela Avenue, Kingsport, Tennessee. 106

Q. What is your occupation, Doctor? A. Chemist.

Q. Are you employed by anyone? A. I am employed by the Tennessee Eastman Corporation.

Q. How long have you been employed by them? A. Since May of 1934.

Q. What has been the extent of your academic training? A. I received the degree of Ph.D. in organic chemistry at Cornell in 1934.

Q. Have you had anything to do with any compounds known as aminophenols? A. Yes.

Q. Over what period has that extended? A. Since October, 1936.

Q. Has your work since 1936 been more or less connected with compounds of that type? A. Yes. Since 1936 a great deal of my time has been spent in work and thought about this type of compound.

Q. Just what is your position at Tennessee Eastman Corporation? A. I am assistant superintendent of the Hydro-Quinone Division of the Tennessee Eastman Corporation.

Q. Do they prepare and market any aminophenol compounds? A. Yes. We manufacture a gasoline inhibitor, one of the constituents of which is nitrogen normal primary butyl para aminophenol.

Q. Are you familiar with the certified copy of Rogers and Voorhees application 427,416 which was filed February 10,
107 1930? A. I am.

Q. Will you please take that certified copy and tell us whether you find in there the name "butyl aminophenol", and, if so, where? A. Yes. On page 8 of this paper, the second line from the bottom reads:

> "butyl, isopropyl and benzyl aminophenols."

Q. Do you find in this certified application in any other place therein the appearance of the name "butyl aminophenol" or any variation of that name? A. No; I don't.

Q. Will you read into the record, please, the entire sentence that you have pointed out on page 8? A. This application is speaking about organic inhibiting agents or antioxidants, and it says:

> "As examples of such materials, we employ the aminophenols, para- and ortho-methylaminophenol, dimethylaminophenol, hydroxyazobenzine, butyl, isopropyl and benzyl aminophenols, dihydroxy diphenyl amine, paraphenetidine, amino naphthol and the like."

Q. As a skilled chemist, Dr. Davy, with a considerable amount of training, when you see in print the term "butyl aminophenol", as you see it on the blackboard here, what does that convey to you? A. What the name "butyl aminophenol" conveys to me depends upon the circumstances of the moment. The words set apart there describe or name a group of compounds which might be called substituted aminophenols in which the substituent group is butyl. That, I
108 think, is all that the name itself denotes.

Q. Does it tell you anything about the type of butyl radical that is included in that name? A. As such it does not.

Q. How many types of butyl radicals are there, Doctor? A. There are four.

Q. Could you name those, please? A. There is the normal primary butyl, the normal secondary butyl, the isobutyl and the tertiary butyl.

Q. As you see that name there (indicating on blackboard) does it tell you anything about where the butyl radical is linked with the rest of the compound? A. No.

Q. Might it be, for instance, a substituent of the phenol group? A. It could not be a substituent of the phenolic hydroxyl group, because that would make it an ether. It could make it a substituent of the benzene nucleus of the phenol.

Q. That is what I mean; around this nucleus somewhere (indicating)? A. That is right; anywhere in that ring not occupied, of course, by some other substituent.

Q. Or it could be hooked up to nitrogen, could it? A. Yes.

Q. With those possibilities, how many compounds might there be under this broad name "butyl aminophenol"? A. May I step to the blackboard and set this forth?

The Court: Yes.

(The witness went to the blackboard and continued his answer as follows:)

The Witness: The name "aminophenol" means a benzene ring in which there are a hydroxyl group and an amino group. So that substituted in the ring these may go into the ortho positions, which are the adjacent positions, or they go into the meta positions, which are adjacent but one, or the para positions, which are separated by two. So the name "aminophenol" in itself includes three possible parent substances.

The name "butyl" merely denotes an aliphatic radical made up of four carbon atoms, of which there are four possible different arrangements.

To take the substitutes in the ring, if the aminophenol is an ortho-aminophenol the butyl group may be substituted in this position (indicating) or in that position (indicating). These two positions are equivalent to those two (indicating). So that you may have any one of four butyl groups in two positions here; that is, eight possible compounds. There are more than eight, because these two groups are different (indicating). Take ortho-aminophenol. The butyl group can have any one of four in here (indicating), any one of four in there (indicating), any one of four in here (indicating), and any one of four in there (indicating). That makes it sixteen.

If these are in the meta position with regard to each other, we may have any one of four here (indicating), any one of four in that position (indicating), any one of four in this position (indicating), or any one of four in this position (indicating). And that is sixteen more.

110 If they are para, if para aminophenol is the parent substance, we may have any one of four in this position (indicating), or any one of four in that position (indicating), and these two are equivalent to those two (indicating); so we may have eight.

Then if the amine group is substituted it is possible to have any one of four substituents of the four butyl radicals there, which will give a total, then, of 44 possible different chemical compounds which are included under the generic name of butyl aminophenols as the word is used in the certified copy of the application which you just handed me.

Q. Is there anything in the certified Rogers and Voorhees application of 1930 which tells you which of those 44 possibilities he had in mind when he used that broad term? A. In reading the context, particularly that full sentence that was just introduced into the record there is nothing to indicate which one or more of the possible 44 is meant. In fact, it can very well be interpreted to mean all 44 butyl aminophenols.

Q. When you say there are 44 possibilities there and that the name "butyl aminophenol" may mean any one or all 44 of them, does that name tell you anything about the properties of the compounds? A. It tells a certain amount to one who is familiar with the aminophenols and their chemical properties. That is, that a part of the group would be expected to be soluble or to form salts with strong alkalis or with strong acids due to the presence of the phenolic hydroxyl and the amino group; but as regards the physical properties or other properties it conveys nothing.

Q. Does it tell you anything about the color of the compound? A. No. I think it might be expected to be colorless. But there is nothing definite; no. 111

Q. Does it tell you anything about its melting point or boiling point. A. No.

Q. Does that name tell you anything about its properties toward other chemicals or compounds other than the broad fact that it is a phenol? A. It says that it will possess in general the chemical reactivity of a phenol and an aminophenol, but nothing such as the derivatives of the amino group and the hydroxyl group.

Q. Does the name "butyl aminophenol" tell you, speaking as of the date in 1930, how to prepare that compound? A. Not at all.

Q. In 1930 was there known in the literature any method of preparation of any butyl aminophenol? A. No.

Q. Did you yourself know any in 1930? A. No.

Q. Had anyone in 1930 ever told you of any such method? A. No.

Q. I wish you would look, Doctor, at the Rogers and Voorhees patent of 1937, of which you have a copy there. A. What number is that, please?

Q. 2,084,977. A. I have it.

Q. That application was filed March 26, 1937, and the patent was issued on June 22, 1937. A. This copy I have does not have the 1937 complete on the patent. 112

Q. I notice that at the end of the specification it says that the application is a continuation "of part of our co-pending application Serial 427,416, filed February 10, 1930." Is that the same serial number as the certified copy that you have been looking at? A. It is.

Q. Now, will you tell us wherein this Rogers and Voorhees patent is equal to the 1930 application and where it differs? Where does it supplement anything? What is disclosed in the 1930 application? A. Well, in this published patent the

113 subject matter is the same in general, that is, directed toward the art of preventing a deterioration of gasoline; and the original application, or this application here, Serial No. 427,416, as we have just said, mentions in a general way butyl aminophenols. The Rogers and Voorhees patent No. 2,084,977 mentions or it specifies the four possible isomeric butyl groups, namely: "the normal butyl radical. The corresponding secondary butyl, tertiary butyl and iso-butyl aminophenols." This patent is specific, whereas application No. 427,416 speaks of these 44 compounds only in a general way.

Q. Is there anything else in this patent more clearly limiting the compounds that the patent is talking about over what are referred to in the application? A. Yes. This application here describes or sets forth the use of a considerable number of compounds, as I read from page 8; aminophenols in general, methyl aminophenol, dimethyl aminophenol, then hydroxyazobenzine, which is not an aminophenol; then the butyl, isopropyl, and benzyl aminophenols; and dihydroxy diphenyl amine, paraphenetidine, which is an ether of an aminophenol, an aminonaphthol; whereas the U. S. patent 2,084,977 relates to the use of the butyl aminophenols only.

Q. And you say there may be one of the four butyl radicals in that compound, according to this patent? A. According to this patent—this patent specifically mentions all of the four possible butyl radicals.

Q. Look at line 3 in the second column; 3, 4, and 5 there.
114 A. Yes.

Q. What does he mean by that statement? A. He says, or they say, "Our present invention relates to the use of ortho-, meta-, and para-butyl aminophenols as gum inhibitors for cracked gasolines."

Q. Does that further limit the compounds he is talking about in this issued patent? A. Yes. When the term ortho- or meta- or para-aminophenol is used, it means or it implies that the butyl radical must be substituted on the nitrogen, because if it were substituted in the ring the terminology ortho-, meta-, and para- would not be used. You would have three substituents there, and those terms do not apply to more than two substituents.

Q. Well, as far as the use of amines is concerned, might we say that this Rogers and Voorhees issued patent boils the thing down quite a bit? A. Oh, indeed it does.

Q. Now, does this issued patent, however, give any method of preparation for the butyl aminophenols that he mentions? A. No.

Q. Are you familiar with a British patent 350,438 which is cited by the defendant in these two actions? A. Yes. I have a copy of it, a photostatic copy, here.

Q. Does that bear any similarity to the Rogers and Voorhees 1930 application about which you have testified? A. Yes, it does. I have read them both for the purpose of comparison, and except for minor changes in the British patent 350,438 it corresponds almost exactly with this abandoned application of Rogers and Voorhees. They were evidently written by the same person, and then one was revised after 115 the other had been written, but in subject matter they are identical, and they are very similar.

Q. Now, at what place in this British patent, if any, does it refer to butyl aminophenol? A. On page 2, column 2, in lines 103 and 104, it says, "Butyl, isopropyl, and benzyl aminophenols," which is the same phrase that occurs here.

Q. Does the sentence or any of the context of this British patent tell you whether he is referring to an ortho-, meta-, or para-amino compound? When I say "he" I mean the British patent; it is in the name of Wade. A. In the case of butyl, no.

Q. What do the words para- and ortho- at line 102 apply to there? A. Well, line 102 reads "para- and ortho-methyl-aminophenol". The ortho and para, being hyphenated, apply to methyl-aminophenol, of course.

Q. I notice in one of the decisions of the Board of Appeals that they speak of a butyl para-aminophenol being disclosed in this British patent. Do you find any such disclosure in this patent? A. I have been unable to find any such.

Q. Now, speaking as of 1930, or in fact any time from 1930 to March, 1934, and referring to any literature or art that you are aware of, is there anything about this British patent that tells you how to prepare butyl aminophenol? A. No; until Bean's latest invention there is nothing in the literature about any method of preparing any butyl aminophenol.

116 Q. Do chemists who know about one compound sometimes theorize on the existence of other compounds which might have some relation to the one about which they do know something? A. Yes, of course. Organic chemistry has grown by that very theorizing; all organic chemists delight in doing that.

Q. But you testified, I believe, that, when you saw a name, that didn't necessarily tell you how to prepare it, didn't you? A. That's right.

Q. Well, can you explain in any way the statement that appears in this British patent where he refers to butyl aminophenols? A. Well, if an organic chemist is thinking about aminophenols and mentions methyl, and he wants to name a large number of them, why, of course the methyl or the ethyl or the propyl or the butyl or phenyl or any of these groups which are known to him are possible as substituents in the parent substance.

Q. Well, is there anything in this British patent, or in fact in the Rogers and Voorhees abandoned application of 1930, which tells you that they had any butyl aminophenol in their hands or in their laboratory? A. No, none whatever.

Q. What do you interpret the statement commencing at line 10 in the Rogers and Voorhees issued patent to mean, the sentence which starts on line 10, column 2? A. Line 10, column 2: "In addition, the benzene ring of the compound may be substituted with hydrocarbon radicals such as the methyl, ethyl and propyl radicals." I think that—well, it means that
117 the preceding substitution was intended to be on the nitrogen, because he distinguishes by saying, "In addition, the benzene ring may be substituted," but it is another example of his theorizing that we have mentioned. The benzene ring

may be substituted, and he names methyl, ethyl, and propyl as well known groups which could be substituted in the ring.

Q. Now, are you familiar with any names in the organic chemistry art that are applied to compounds which do not exist, other than what you have said about butyl aminophenol as of 1930? A. Oh, yes. After all, the systematic name of a hypothetical compound can be used even though the compound is not known or in fact is incapable of existence. A systematic name merely represents in a shortened form the structural formulas such as are on the blackboard there (indicating) and, as such, merely represent a hypothetical number and kind and arrangement of atoms in a theoretical molecule which may or may not have existence of fact.

For example, a name which is commonly used and commonly spoken of in organic chemistry is vinyl alcohol. Derivatives of vinyl alcohol are well known. Vinyl acetate is used in manufacturing plastics. Vinyl chloride is known, but vinyl alcohol is unknown, and all methods of synthesis which might be expected to lead to the production of vinyl alcohol lead not to vinyl alcohol but to acetaldehyde, which is an isomer.

(The witness went to the blackboard.)

I could explain that more fully. The name "vinyl alcohol," when used either orally or written, means, to an organic chemist, this formula: 118

(The witness wrote on the board as follows:

$CH_2 = CH — OH$.)

If the compound is known, it also means the compound. If the compound is unknown, it means the formula.

Now, this formula does not represent any molecule which has physical existence. At least, it has been impossible to demonstrate that it has. But, rather, if this hydrogen atom in this formula were shifted to this carbon atom (indicating), which would necessitate a shifting of this double bond to between the carbon and oxygen (indicating), we would have this formula:

(The witness wrote on the board as follows:

$CH_3 — CH = O.)$
which is the formula for a known compound, known as acetaldehyde.

All attempts to synthesize vinyl alcohol or all reactions which might be expected to lead to the production of vinyl alcohol lead to the production of acetaldehyde; and, even though derivatives of vinyl alcohol are known, vinyl alcohol itself is not known, and the name "vinyl alcohol" means only this formula (indicating).

Now, there are others, like ortho-formic acid, or 1, 2, 3, cyclo hexatriene, and the 1, 2, 4 cyclo hexatriene. I mean any chemist who put his mind to it could extend the examples almost without limit.

Q. Are you familiar, Doctor, with the Clifford patent No. 2,109,164? A. I am.

119 Q. Would you tell the Court, please, what is the general import of this Clifford patent? A. Yes. The title of the patent is, "Rubber Composition and Method of Preserving Rubber." The patentee is an employee—assigns his patent to the Wingfoot Corporation, which is a manufacturer of rubber, rubber articles. The tests that are shown, the preservatives which Clifford mentions, are all directed to the preservation of rubber and their efficacy as preservatives of rubber. There are only two claims, both of which are directed to the art of preserving rubber.

He mentions, of course, in the opening paragraph here, not only rubber; he mentions deteriorable organic substances and balata and gutta percha, transformer oils, gasoline, and other deteriorable organic compositions, but his whole attention is devoted to the art of preserving rubber.

Q. I notice quite a number of compounds set forth in this patent, Doctor. Have you counted approximately how many compounds are mentioned in this patent? A. Yes; there are approximately 150 to 160 compounds—well, I think you are dignifying these names unduly when you call them compounds. There are, I would say, 155 to 160 names in the patent.

Q. Well, now, just generally looking over those—or I presume you have—what is the general impression you get from this long list of compounds commencing on page 1 and continuing into pages 2 and 3? A. It is hard to get any clear impression of what Clifford was driving at. So many of his compounds or of his names here which are purported to be compounds are, well, classes or groups of compounds. The names could mean two or three or four or more possible compounds, and some of the names just don't have any meaning, under any system of nomenclature that I have ever heard of.

* * * *

Q. Where in this Clifford patent, if any place—you have another copy of it there, do you? A. I have one here, yes. 121

Q. Where do you find in this patent any description of—well, let us say any reference, first, to any butyl aminophenol? A. In line 7, or line 8 of column 2 of page 2, he says, "Also, nitrogen-butyl para-amino phenol," and then in line 11 he says, "nitrogen-isobutyl para-amino phenol," and in line 18 of column two of page 3 he says, "normal-butyl para-amino phenol," but I understand that is a misprint and that he means nitrogen. Isn't that what you said?

Mr. Moore: That is agreed, I believe, that the copy of the—

The Court: (Interposing) The small n should be large N.

Mr. Moore: Yes. 122

The Witness. All right. Well, in that case it is the same reference—it is the same name as appears on page 2.

By Mr. Mayne:

Q. He says there that it can be prepared reacting butyl amine and para-chloro phenol in an autoclave. A. Yes.

Q. Did you ever try that process out, Doctor? A. Yes.

Q. Just briefly describe what you did and when you did it. A. Let's see. I believe in May of this year. I placed three moles of normal butyl amine and a mole of para-chloro phenol in a stainless steel autoclave and heated the contents of the autoclave to 200 degrees, 210 degrees—in that order—for approximately eight hours. I believe that the autoclave was allowed to stand overnight; it came at the end of a working day. And in the morning it was cool, and the autoclave was opened, and an excess of caustic was added, and the butyl amine distilled away—the excess butyl amine, which is unreacted butyl amine—and leaving behind then an alkaline solution, the para-chloro phenol, and any N-butyl para-aminophenol which might have been formed. I then made that alkaline solution strongly acid with hydrochloric acid, which released the para-chloro phenol and converted any possible aminophenol into its acid salt. I used hydrochloric acid. Then I extracted the unused para-chloro phenol from the solution with diethyl ether and tested the extracted solution with sodium nitrite in order to ascertain whether or not any butyl aminophenol was present. The test was negative.

123 Then to convince myself that the test was a valid test, I took a portion of this acid solution and added to it an amount of authentic butyl aminophenol which would be equivalent to a one percent yield: that is, one percent of the theoretically possible yield. Then to this solution containing the authentic butyl aminophenol, the acidic solution, I added a dilute solution of sodium nitrite and got the immediate precipitation of a brown, oily material in considerable quantity. I would estimate that I could have observed in the previous test as little as a tenth as much or less. And these tests showed, then, that the yield of the process that I carried out was certainly no greater than one tenth of one percent of the theoretical quantity, if any at all.

Q. Is that test with sodium nitrite sometimes called the nitroso test? A. Yes, that is the nitroso test, the standard test that has been mentioned on the stand here before.

Q. Then, your net result from trying Clifford's process was zero, I gather? A. Well, yes. And I tried also using cuprous chloride as a catalyst. That is a catalyst that is used for converting chloro benzene to aniline, and it is generally known as a catalyst for this reaction; and carried out the reaction substantially as before, except in the presence of cuprous chloride as a catalyst, and with the same negative results. And I also attempted the reaction of the isobutyl amine and para-chloro phenol, with the same negative result. I might say, it wasn't unexpected. It is commonly known that the chlorine in para-chloro phenol is not very reactive in this 124
type of reaction; that, while it can be made to react with caustic to form hydroquinone, perhaps, with amines it doesn't follow—with the higher molecular-weight amines it apparently does not act as it does with the stronger amines.

Q. I notice in your first example of that preparation or attempted preparation you used three moles, three molecular weights of the butyl amine to one of the para-chloro phenol. Why did you use three instead of two or one? A. Well, it is quite common, if you want a reaction to go, to increase the amount of one reagent in order to take advantage of the mass effect. In other words, if the reaction went it was to be expected that it would go more rapidly and more completely in the presence of an excess butyl amine than it would in merely the theoretical quantity required for the reaction. It is a standard practice, if you are anxious to get a reaction to go, to provide as favorable conditions as possible by using an excess of one reagent.

Q. Now, at the time you made, at any rate, your first run on the Clifford patent and selected the conditions which you have described that you used, were you aware of anyone else making runs to test out the operability of the process in the Clifford patent? A. No, I don't believe so. I think our management had a letter from you asking us to see whether that reaction would go, and I was given the job of trying to make it this way. I mean, after all, we make this normal butyl para-aminophenol and sell it; and, if this reaction is good and if you can make it this way, we certainly want to know about it. That was in our minds at the time.

125 Q. And what you have described is your own interpretation on your way to try to operate that process; is that correct? A. Yes, I tried to operate it.

Q. I have a report here dated October 10, 1941. Can you identify the signature on that report? A. Yes, that is my signature.

Q. Is that a correct description of your first runs with the Clifford process? A. Yes.

Q. Was that compiled from your notebooks recently? A. Yes, that was compiled from my notebooks. I think it is a transcript of my notebook, really .

> Mr. Mayne: I should like to offer this report in evidence as Plaintiff's Exhibit No. 21.

> (Typewritten report on investigations of Clifford U. S. Patent 2,109,164, October 10, 1941, was marked Plaintiff's Exhibit 21 and received in evidence.)

* * * *

132 Q. Is the word "para hydroxy diphenyl amine," which is used at lines 62 and 63 of column 2 on page 2 of this Clifford patent—is that name a synonym for butyl aminophenol, Doctor? A. No, no. No.

Q. They are not the same compounds, then? A. No.

> Mr. Mayne: I believe that closes this gentleman's testimony, your Honor, as to the direct.

Cross Examination by Mr. Moore:

Q. Dr. Davy, are you a laboratory man or are you a businessman with the Eastman Kodak Company at Kingsport? A. Well, partly both. I mean my duties are partly—well, when I came to the Tennessee Eastman I went in the Research Department, did laboratory work. Then I was trans-
133 ferred to the Hydroquinone Division. At the present time I do laboratory work with a helper and am responsible for

the direction of all the experimental work that is done in our division of the company; and then on the side or in addition I have some administrative duties.

Q. There was great interest in aminophenols in 1936, wasn't there, as antioxidants for gasoline? A. Yes.

Q. That is when you started your operations? A. Yes, we started the manufacture and sale of them, I believe, in 1936.

134 Q. In 1936 there was a time when a series of patents were issued by the Patent Office on aminophenols broadly, containing broad claims; is that right? A. I am not sure. Our experimental work on these compounds was going along, and since 1934 we have been trying to manufacture these compounds; but I am not sure about the dates as to when they were actually put on the market or when the patents were issued.

Q. You do remember in 1936 that your competitors were quite active in aminophenols as gasoline antioxidants, do you not? A. If you say so. I am not sure.

Q. Now, refer to the Rogers patent which I presume you have before you. That is patent 2,084,977. A. I have it here.

Q. I will drop that question. Just refer to the Wade British patent 350,438, and turn to page 2 of that patent, the second column. A. Yes.

Q. In line 100, the sentence beginning in that line. Is it not a reasonable construction of that sentence to have the "para" modify the "butyl", and hence call for para butyl aminophenol? A. No. I think that sticking that "dimethylaminophenol" in there, where you see "ortho" and "para", makes all the rest in the general thereafter. If you read it all, you will see it says "para- and ortho-methyl-aminophenol, dimethyl-aminophenol, butyl, isopropyl and benzyl aminophenols, dihydroxy-diphenyl-amine, and the like."

135 Q. Butyl aminophenol is definitely specified, is it not? A. Phenols; yes.

Q. And does not that mean the normal aminophenol and not the secondary or tertiary form? A. That depends entirely upon the experience of the chemist. At one time in our company we were handling isobutyl alcohol, two or three hundred pounds a day; and at that time when anybody said "butyl alcohol" it means that to us. At another time we were handling isopropyl, and when anybody said "propyl alcohol" we all said "propyl alcohol", meaning isopropyl. At another time butyl has meant secondary butyl to me. Unless you define the circumstances it is entirely dependent upon so many other factors than those that you say.

Q. Let us look at the context of this sentence. After "butyl" you find "isopropyl," do you not? A. That is right.

Q. Other than that, if the patentee had intended anything more than normal butyl to be specified, he would have said so, inasmuch as he said in the following words "isopropyl" instead of "propyl" broadly? A. Perhaps. Or it may mean that he was working with isopropyl alcohol, or something like that, in that group, and the isopropyl was the one he thought of. If you are going to theorize on what was in the mind of the writer—

> The Court: Counsel has not asked you what was in the mind of the writer. He has just asked you the simple question, When you see that expression there does it not mean ordinary butyl?

The Witness. No, sir.

136 The Court: What does it mean?

The Witness. It leaves unmentioned which butyl is meant. It has quite often been my experience that when the general term "butyl" has been used I have asked whoever was talking just which butyl he meant.

By Mr. Moore:

Q. Do you know of any nitrogen para-aminophenol that will not operate in some degree as an antioxidant? A. Do you mean, potent.

Q. In any degree, I said. A. Cyclohexyl is so insoluble that you cannot get enough in solution for it to be an anti-oxidant.

Q. If you can get such a compound in solution, do you know of any one that will not operate in some degree as an anti-oxidant? A. No; I think not.

Q. Look at the Clifford patent, will you, please? You say there are several hundred compounds mentioned by Clifford. Are they not all aminophenols or chemically related compounds? A. No; they are not.

Q. How many are not? A. I have not gone over them with that in mind. He mentions a number of quinones. He mentions a number of nitroso bodies. He gives a general formula which could include, primary, secondary, and tertiary amines. That is what he mentions here. He mentions a much broader group of compounds than merely aminophenols?

Q. The aminophenols or derivatives of the aminophenols? A. I have not studied it from that point of view. 137

Q. Referring to the general method set forth by Clifford at the bottom of Column 2 of Page 2, you did not actually try the method he referred to there, using butyl amine, hydroquinone and calcium chloride? A. I have not tried it with normal butyl amine. I have tried it with secondary butyl amine, long before I ever saw this patent.

Q. In preparation for the trial of these cases and in connection with your report of October 10, you did not try the method? A. No.

* * * *

139 Q. You have heard of the Wade British patent. Wade definitely says, does he not, that, "As examples of such materials, we have employed"— A. Where is that?

Q. Page 2, line 100. A. Yes.

Q. Clifford specifically says, too, that his compounds are usable as gasoline antioxidants besides being useful in the rubber art? Is not that true? A. Yes. That is a customary statement in a patent. 140

Q. And Clifford, then, in this patent, in addition to mentioning butyl para aminophenol, refers on page 3 to the proposed method of preparation of that compound, does he not? A. Yes.

Q. And he says on page 1, column 1, lines 7 to 10:

> "The substances to which the invention relates have been found by actual test to protect against deterioration from such influences as heat, light and oxygen"?

A. Yes. He does not specify what deteriorable substances he has tested, however.

Q. When you ran these tests in May, 1941, there was no representative from the Patent Office present, was there? A. No.

Mr. Moore: I believe that is all.

By the Court:

Q. As to Defendant's Exhibit No. 20, you said you underscored the Clifford patent. You said you underscored on page 2 the various matters there that could not be produced or were not practicable? A. No, sir. I first went over the list. The patent is so vague in the naming of compounds, and I was asked if there were any vague and uncertain names in the patent, and I underlined those names, and in doing so I found a number which have no meaning.

Q. And that is what you meant by "underscored"? A. Yes, sir.

Q. Will you look on page 2, column 2, line 8? It reads: "Also, N-"— That stands for nitrogen, does it not? A. Yes,
141 sir.

Q. (Continuing reading): "N-butyl p-amino phenol." You say that that term is vague, that it does not exist? A. No, sir.

Q. You underscored it? A. Yes. By underscoring it I meant that more than one compound may exist which could be included under that name.

Q. You say it is used as an expression of some compound that does not exist? A. No, sir. I have not made my point clear, evidently.

* * * *

Re-direct Examination by Mr. Mayne: 145

Q. I should like to ask you a question or two in regard to this matter of nomenclature. I inquired for the so-called radical index of the American Chemical Society yesterday. Have you gone to the library since you were on the stand and examined that radical index? A. Yes, I have.

Q. Will you explain whether that would affect your answer with respect to the cross-examination question having to do with amyl aminophenol? A. Having a radical index 146 which demonstrates to the reader how the names in the index are meant to be construed, there can be no question but that when amyl is used in the index of chemical abstracts it means the normal primary amyl that is shown in the 1937 volume, which contains a list of the names of organic radicals.

Q. Now, is this abstract index followed universally by American chemists or not? A. No, it is not. The chemical abstract list is compiled for the purpose of indexing, and of course a very weighty consideration is brevity and economy. There is a considerable effort to have the system universally used, but that effort has not been entirely successful by any means.

Q. Now, if you follow the chemical abstract system would the name "nitrogen butyl para-aminophenol," which you see at line 8, column 2, page 2 of the Clifford patent be a specific or a general name? A. It would be a specific name. There can be no doubt, if the system that is used to index chemical abstracts is followed, that nitrogen butyl para-aminophenol means nitrogen normal primary butyl para-aminophenol, and that name should not be underlined. Also, if the system the chemical abstract uses for indexing is followed, in line 11 "nitrogen-amyl para-amino catechol" might not be underlined.

Q. Is there anything in the Clifford patent to indicate to you whether he followed the American abstract system or

whether he followed some system of his own, let us say? A. He makes no statement that he follows the chemical abstract
147 system for indexing, and as far as the large majority of the names go there is no way of telling whether or not he was following, except from the general ambiguity. However, on page 3 of the patent there are a number of formulas, and where he puts forth these formulas and gives the names for the formulas he means that the names shall be equivalent to the formula.

Now, in column 1 on page 3, in the space between lines 42 and 53, he has a formula. The formula leaves no ambiguity whatsoever, but the name which he says is equivalent to that formula does not follow the chemical abstract's indexing system. He gives the formula—perhaps I could better set this forth on the blackboard.

Q. I think his Honor has a copy of the patent there; he can see that.

The Court: I can follow it.

By Mr. Mayne:

Q. That is the Clifford patent. A. He says, "2-hydroxy 1-4-anilido naphthoquinone," comma, and then gives the formula. Now, the name "2-hydroxy 1-4-anilido naphthoquinone" doesn't really make sense. He either should say, well, "1-4-di-anilido naphthoquinone"—the name without the formula doesn't convey anything to me, and I don't think it conveys anything to 'most anybody except Clifford. The proper name to be used if you are following the chemical abstract's indexing system would be 2-hydroxy 1-4-naphthoquinone 4-phenyl imide.

Now, at the bottom of the page he has another formula in which he says, "the compound 2-hydroxy para toluidino 1-4-naphthoquinone having the formula," which he gives. The
148 same remarks would apply. That name is not in the chemical abstract's indexing system. The correct name would be 2-hydroxy 1-4-naphthoquinone 4-para-methyl phenyl imide, or 4-para-toluimide.

And then there is another name in lines 74 and 75, and at the top of column 3 another formula and name. These would indicate to me—I mean, just reading the patent by itself—that either Clifford was unfamiliar with the chemical abstract system of indexing or, well, at any rate he didn't use it.

Q. You testified that you acted as assistant superintendent of the hydroquinone division of Tennessee Eastman Corporation, I believe? A. That is correct.

Q. In addition to your research duties? A. That is correct.

Q. Did Tennessee Eastman Corporation ever market a gasoline antioxidant containing as the active constituent nitrogen normal primary butyl para aminophenol? A. Yes, we have been manufacturing and selling an inhibitor with that as the principal active constituent since 1936.

Q. And what is your trade name for that inhibitor, Doctor?

A. It is marketed as UOP-4. On our records we carry it as TE-4.

Q. You sell this inhibitor, then, to the Universal Oil Products Company? A. Yes.

Q. And you started in 1936, you say? A. To the best of my memory, yes.

Q. Have the sales since 1936 increased at all, or has there just been a minor interest in it? A. Well, in 1936 as we introduced it it started out in small quantities, and the volume of sales has been increasing until now we are manufacturing and selling several hundred thousand pound per year. 149

Mr. Mayne: That concludes his testimony, your Honor.

Re-cross Examination by Mr. Moore:

Q. Dr. Davy, the system adopted by the American Chemical Society in the radical index didn't overturn the existing system of nomenclature, did it? A. No. Rather, I would say that it codified it. There were so many possible ways of naming compounds that ambiguities are all through the literature, and the purpose of the classification which was initiated by the American Chemical Society and which represents a study of several years was found to be necessary because of the fact that chemists are liable to be ambiguous in naming terms.

Q. Isn't this nitrogen normal primary butyl para aminophenol sold as an antioxidant to only one customer, the Universal Oil Products Company? A. Well, as I understand it, the Universal Oil Company, through their connections with the oil refining industry, acts as our sales agent in effect.

Q. Your agent. Your sales are to Universal Oil Products? A. No. Our shipments are made direct from Kingsport, Tennessee, to, oh, the Texas people and the Shell people and to Standard, out to as far as the Bahrein Islands. We ship direct from Kingsport to the refiner. 150

Q. Through sales made by U. O. P.? A. That is correct.

Mr. Moore: That is all.

* * * *

ALBERT COUCH RUGGLES was called as a witness for and on behalf of the plaintiff and, being first duly sworn, was examined and testified as follows:

* * * *

156 Cross Examination by Mr. Moore:

Q. Dr. Ruggles, you find no specific reference in so many words in this report of yours to nitrogen normal primary butyl para-aminophenol, do you? My copy of it, your report (indicating).

(There was no response.)

Q. Do you have the question? A. No, I don't understand the question.

Q. Do you find in so many words a reference in that report to nitrogen normal primary butyl para-aminophenol? A. No, I do not.

Q. You have used in that report the term "nitrogen butyl para-aminophenol," haven't you? A. Yes.

Q. You are referring to the same compound in that report that you refer to on the stand, are you not? A. Yes. 157

Q. Were you responsible for the nomenclature used in this report? A. Yes.

Q. Now, you say that one sample tested, the sample that is reported on the bottom of the page, was made in Kingsport. Did you prepare that sample? A. Yes.

Q. By what method? A. By the reaction of hydroquinone and normal primary butyl amine.

Q. Where did you receive the information that enabled you to prepare that? A. It was the result of previous experimental work.

Q. By you? A. Yes, by me and others.

Q. The tests which you have referred to and reported on page 11 of the application serial No. 134,903 show, do they not, that nitrogen 2-ethyl normal primary butyl para-aminophenol was a poorer gum inhibitor than nitrogen normal primary butyl para-aminophenol itself? A. Yes, that is correct.

* * * *

CHARLES G. DRYER was called as a witness on behalf of the plaintiff and, being first duly sworn, was examined and testified as follows: 158

DIRECT EXAMINATION

The Assistant Clerk. State your full name, please.

The Witness. Charles G. Dryer, D-r-y-e-r.

By Mr. Mayne:

Q. Where do you reside, Mr. Dryer? A. 4135 Ellington Avenue, Western Springs, Illinois.

Q. And what is your occupation, Mr. Dryer? A. I am a chemist and have been closely associated with work in connection with the treatment of motor fuels and the application of antioxidants to gasolines.

Q. Are you employed by anyone at the present time? A. Employed by the Universal Oil Products Company.

Q. Their offices are at Chicago, I believe? A. Chicago.

Q. And what is the nature of your academic training, Mr. Dryer? You are a graduate chemist, are you?

A. Yes, sir. I have a Master of Science degree from Purdue University in organic chemistry.

Q. Now, your duties, you say, at Universal Oil Products Company are in connection with antioxidants? A. Yes, sir.

Q. Have you been connected with Universal Oil Products long? A. I have been employed since 1930.

Q. And have your duties been more or less along that line 159 throughout that entire period of employment? A. I have either been actively engaged in this type of work or in a supervisory capacity of a research group that has had this as their main project.

Q. And does your work bring you in contact with the industry in any way, Mr. Dryer? A. Yes, sir, it does. The petroleum industry in general we are in quite close touch with, both in connection with the treatment of motor fuels in general as well as the direct application of antioxidants to the particular fuels that are made generally throughout the entire industry.

Q. Why does the industry feel, if you can tell us, that they have to use an antioxidant in gasoline? A. Unrefined motor fuels that are normally produced by the various cracking processes undergo oxidation reactions which result in the formation of gummy or resinous materials unless suitable methods for the treatment of these fuels are used or unless antioxidants are employed subsequent to the manufacture of these fuels. The gum content of a fuel has a direct effect upon the operation of any internal-combustion engine, and it is important and essential that the gum content of fuels be maintained at a low figure.

Q. In the practical operation of a gasoline engine or motor what would happen if you didn't inhibit the gum formation in the gasoline? A. If the gum content was not retarded in some way, the gasoline would form sufficient gum to actually stop the operation of the motor, as has been observed in many cases.

Q. Has your research division that you are supervising tested very many antioxidants: that is, just a few, or have 160 they tested quite a number? A. I should say we have tested a large number.

Q. As a result of your work what would you say, Mr. Dryer, are the requirements for a good gasoline antioxidant? A. Well, in addition to having a high effectiveness, an antioxidant should be readily soluble in hydrocarbons such as motor fuels, insoluble to water to a large extent; they should impart no color upon addition to the gasoline; they should have no offensive odors; and they should likewise have no effect on the dyes or any anti-knock material, such as tetra ethyl lead, that is added to the motor fuel.

Q. Do you ever find, in your experience, a compound occasionally that possibly has good inhibiting value itself but is ruled out because of water solubility, for instance? A. Oh, there are many compounds that are very effective with respect to stabilizing a fuel, but from a practical standpoint they are ruled out because of other physical properties, of which water solubility may be one.

Q. Have you ever tested nitrogenmethyl para-aminophenol, for instance, as an antioxidant? A. Yes, sir.

Q. Does it have good inhibiting value per se? A. Yes, sir.

Q. How about the water solubility? A. Its water solubility is high, and it is difficulty soluble in hydrocarbons.

Q. And would you regard that as a good commercial antioxidant? A. No, sir.

Q. Now, just tell us, please, how this water solubility comes 161 into play with commercial antioxidants. Why are we concerned with that? A. In the refining of motor fuels they are frequently given many other methods of treatment which require water washing following whatever treatment the fuel may be given, and in the majority of cases it is quite difficult to produce a fuel that is entirely free from water, and the inhibitor is added on some occasions to a gasoline that contains very small traces of water. As a result water accumu- 162
lates in the storage tanks in which the fuels are stored and,

as a result, antioxidants that are water-soluble are leached from the motor fuels under those conditions, rendering the gasoline in a condition whereby it can continue to oxidize and form gummy materials in the absence of the added inhibitor.

Q. In your trips around through the industry what do you find may be the average maximum storage period for gasoline? May it be held in storage more than a few days or weeks? A. It depends largely on the season of the year and also the demand; but it will vary up to, in many cases, as long as a year's storage. That is, I should say, in the past several years.

Q. And there would be ample chance for water accumulation in the storage, would there? A. Yes.

Q. How does this color requirement enter into the properties of an antioxidant? A. Well, the color of a motor fuel as it is normally marketed—some are marketed as water-white fuels, but the majority of them are marketed as dyed fuels and have a distinctive color. If an antioxidant imparts color to these fuels it detracts from the sale of the product in the water-white class and it also changes the color of a dyed fuel.

Q. In connection with or related to that color situation, is there ever a situation where there is a precipitating out that occurs in gasoline? A. Under some conditions. It is more noticeable, I should say, under exposure to light where precipitation actually occurs. 163

Q. Then an antioxidant must have some light-stability properties, must it not? A. Yes, sir.

Q. In view of all these requirements for a good all-around gasoline antioxidant, do you think a man skilled in the art is able to predict what antioxidant to use? A. I should say, generally speaking, that the predictability of the general properties of an antioxidant is quite difficult and could probably not be done with any degree of accuracy.

Q. Then, if you see the name of a compound any place in the literature, you would not be able to say much about its antioxidant properties, I take it? A. If it was a new compound and I knew nothing of its properties; no.

Q. Have you ever seen the certified copy of the abandoned Rogers and Voorhees U. S. application 427,416 (handing a paper to the witness)? A. Yes.

Q. On page 8 of that application, near the bottom of the page, I believe, there is a statement about certain compounds that those inventors allege they have used as gasoline antioxidants. Is that correct? A. Yes.

Q. They name several compounds there. Are there any that you have tried and found of doubtful operability? A. I should say that the next-to-the-last line, para-phenetidin—that would be of little utility as a gasoline antioxidant, with 164 properties that would be of no commercial significance whatsoever.

Q. You have actually tried that compound, or your division has, as an antioxidant? A. Yes, sir.

> Mr. Mayne: I would like to offer in evidence as Plaintiff's Exhibit No. 25 the certified copy of the Rogers and Voorhees abandoned application No. 427,416.
>
> (Certified copy of Rogers and Voorhees abandoned application 427,416 was marked Plaintiff's Exhibit No. 25 and received in evidence.)

By Mr. Mayne:

Q. In that same list of compounds at the bottom of page 8 of this application are there any that you can say were on the market prior to the close of 1936?

> Mr. Moore: That is entirely immaterial, if your Honor please.
>
> The Court: How will that help the Court? However, I will let it in. He may answer.

A. Yes, sir.

By Mr. Mayne:

Q. Which one or ones? A. Benzyl aminophenol.

Q. Are there any others that were on the market prior to 1937? A. No; not for use in motor fuels.

Q. Just tell us, then, what has been the history of motor fuel antioxidants that were on the market since you have 165 been connected with this industry from, let us say, 1930 on; which ones have been commercial antioxidants? A. There have been benzyl aminophenol, materials prepared from the distillation of wood products—

Q. Excuse me. Can you just make that sort of chronological from 1930 on, in a rough way? A. In 1930 there was a product prepared from the distillation of wood products; there was benzyl aminophenol, alpha-naphthol, tri-cresol, and nitrogen primary normal butyl para aminophenol.

Q. When did you first hear of the latter compound, namely, the nitrogen normal primary butyl para aminophenol? A. In 1936.

Q. From whom or from what organization did that information come to you? A. We received that information from the Tennessee Eastman Corporation, Kingsport, Tennessee.

Q. Did your company undertake the marketing of a gasoline antioxidant of which that was an active constituent? A. Yes, sir.

Q. When did that occur? A. That took place in the latter part of 1936.

Q. Had benzyl-aminophenol been a prominent antioxidant in the industry prior to that? A. Yes, sir.

Q. What was the general course of the sales of this butyl aminophenol that you have referred to? A. Of course when sales first started they started out in a rather small way, but shortly thereafter the sales increased, and they have shown a 166 steady increase in volume annually, and at the present time there are several hundred thousand pounds marketed annually, of this material.

Q. Did you ever hear of any butyl aminophenol being urged upon the market prior to your receiving information about it in 1936 from the Tennessee Eastman Corporation? A. No, sir.

Q. And you were in almost daily touch with the industry, were you? A. Yes, sir.

Q. Has benzyl aminophenol continued to be a prominent factor in the gasoline industry up to the present time? A. No, sir. It has been largely discontinued in the stabilization of motor fuels in the past two or three years.

Q. Did those who were marketing the benzyl aminophenol as a gasoline antioxidant just quit selling antioxidants, or did they offer a substitute for that benzyl compound? A. No, sir; they offered a substitute for the benzyl compound.

Q. What type of compound did they offer as a substitute? A. The active ingredient was substantially nitrogen primary normal butyl para aminophenol.

Q. About when did that offer first occur by the other people in the industry? A. That became a commercial reality in approximately October of 1938.

Q. That is, with your competitors? A. Yes.

Q. Does Universal Oil Products obtain all of their nitrogen normal primary butyl para aminophenol from the Tennessee 167 Eastman Corporation or elsewhere? A. All of the material that is sold throughout the industry is obtained from the Tennessee East Corporation.

Q. That is sold by the Universal Oil Products? A. That is right.

Q. And you market that under what trade name or number? A. It is marketed as U. O. P. No. 4 Inhibitor.

Q. You deal with the Tennessee Eastman Corporation on the basis of what number? A. As T. E. 4 Inhibitor, to Tennessee Eastman Corporation.

Q. As you have observed inhibitors come and go, let us say, in the last decade, would you regard this T. E. 4 or U. O. P. 4 gasoline inhibitor as a commercial success? A. Yes, sir; very much so.

Q. Was there known, say, a decade ago, ethyl-para-aminophenol as a compound? Did you ever test that compound out, for instance, as an inhibitor? A. No, not a decade ago.

Q. Have you ever tested it out? A. I have tested it out in recent years.

Q. Did that prove to be a good inhibitor for gasoline? A. It was effective, but, due to other properties it was not particularly desirable material for all of the requirements of commercial antioxidants.

Mr. Mayne: That is all of my direct examination.

Cross Examination by Mr. Moore:

168 Q. Doctor, para-phenetidin is not an aminophenol, is it? A. It could be classified, perhaps, as a derivative.

Q. If you saw a reference to normal butyl para aminophenol as an antioxidant you would at least feel it worth a try as a gum-inhibitor for gasoline, would you not? A. I would consider it worth a try.

Mr. Moore: That is all.

Mr. Mayne: That is all.

(The witness left the stand.)

Mr. Moore: Does that close your case, Mr. Mayne?

Mr. Mayne: Unless we have overlooked a possible exhibit about which we have talked. I do not think we have.

Mr. Moore: I believe I offered the von Bramer patent for identification as Defendant's Exhibit No. 3. I want to offer it now as Defendant's Exhibit No. 3.

(Copy of von Bramer patent previously marked for identification as Defendant's Exhibit No. 3 was received in evidence.)

Pltf. Ex. No. 1

SPECIFICATION
of
UNITED STATES PATENT APPLICATION
of
Frederic R. Bean
of
Rochester, New York
for:
"N SUBSTITUTED AMINOPHENOLS AND MOTOR FUELS STABILIZED THEREWITH"

Serial No. 134903 filed April 3, 1937

This invention relates to N-substituted aminophenols and to motor fuels stabilized against deterioration therewith.

It is known that N-aralkylaminophenols can be prepared by treating aminophenols with aralkyl halides. For example, N-benzyl-p-aminophenol can be prepared by reacting p-aminophenol with benzyl chloride. This method of preparing N-aralkylaminophenols suffers from the formation of side-reaction products, e. g., N, N-diaralkylaminophenols. These side-reaction products can be removed only with considerable difficulty. It has been proposed to prepare N-alkylaminophenols in a similar manner from alkyl chlorides and aminophenols. However, such a method requires the use of high temperatures, owing to the lower reactivity of the alkyl chlorides. In addition the yields are poor and the N-alkylaminophenols, if formed, is contaminated with side-reaction products, decomposition products and unreacted aminophenol. Separation of the N-alkylaminophenols is an involved and tedious procedure. It has also been proposed to prepare

N-substituted aminophenols from N-aralkylidene aminophenols or N-alkylidene aminophenols by treatment thereof with alkali and zinc. This method suffices fairly well for the production of N-aralkylaminophenols, but is less satisfactory for the preparation of N-alkylaminophenols.

Now I have found a method for preparing N-substituted aminophenols which gives rise to products of good purity and which is particularly adapted to the preparation of N-alkylaminophenols containing at least two carbon atoms in the alkyl group. By my new process, heretofor unknown N-alkylaminophenols having unusual properties can be made. These new N-alkylaminophenols are useful as photographic developers and are particularly adapted to the manufacture of motor fuels stabilized against deterioration, I have found.

An object of my invention, therefore, is to provide a process for preparing N-substituted aminophenols. A further object is to provide new N-alkylaminophenols. A still further object is to provide motor fuels stabilized against deterioration with such new N-alkylaminophenols and a process for preparing such stabilized motor fuels. Other more specific objects will appear hereinafter.

According to the process of my invention, an aminophenol is reacted with an aldehyde of two or more carbon atoms, in the presence of a strong base and aluminium. The aldehyde probably reacts with the aminophenol to form a compound of the following formula:

$$\text{I.}\quad \text{R—CH=N-phenylene—OH}$$

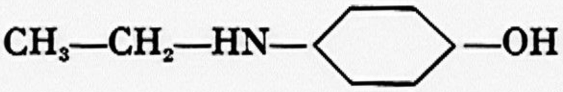
wherein R represents an organic group. (I.) is then reduced immediately by the action of the strong base and aluminium to give a N-substituted aminophenol. In a less preferred embodiment of my invention, a compound of formula (I.) can be used as a starting material. However to obtain higher yields, I have found it necessary to effect the reduction of the intermediate compound as quickly as possible. N-aralkylaminophenols can be prepared by my new process but the process is particularly adapted to the preparation of N-alkylaminophenols from aminophenols and aliphatic aldehydes which form intermediate compounds of the type (I.) but slowly.

While the process of my invention lends itself to variation and modification in the manner of its practical application, particularly as regards the nature of the strong base, the form of the aluminium, the proportions of reacting materials and agents used in the reaction, the temperatures and the exact method of procedure, the following examples will serve to illustrate how the process can be practiced.

Example 1. *N-ethyl-p-aminophenol*

218 g. (2 mol.) of p-aminophenol were dissolved in 5000 cc. of water containing 142 g. (3.5 mol.) of sodium hydroxide. The solution was placed in a vessel equipped with a means of stirring and the air in the vessel was displaced with a non-oxidizing atmosphere, such as hydrogen or nitrogen. The solution was cooled to about 5°C. and 90.5 g. (2 mol. with slight excess) of acetaldehyde were added. 100 g. of activated aluminium were added gradually while cooling and stirring rapidly. After addition of the aluminium, the reaction mixture was stirred rapidly for from two to three hours at about 25°C. The reaction mixture was then made just acid with sulfuric acid. The resulting solution was concentrated to a volume of about 2000 cc. and decolorized with activated charcoal. Upon cooling the decolorized solution of N-ethyl-p-aminophenol sulfate separated out after standing.

Upon dissolving the sulfate in water and making just alkaline with sodium carbonate mixed with a little sodium sulfite, N-ethyl-p-aminophenol separated out in the form of white crystals melting at 111° to 112°C. Its nitroso derivative melts at 81° to 83°C. with decomposition. The formula for N-ethyl-p-aminophenol is:



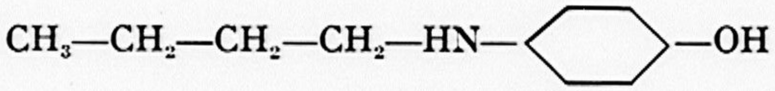
Example 2. *N-(n-primarybutyl)-p-aminophenol*

218 g. (1 mol.) of p-aminophenol were dissolved in 5000 cc.

of water containing 142 g. (3.5 mol.) of sodium hydroxide. The solution was placed in a vessel equipped with a means of stirring and the air in the vessel displaced with a non-oxidizing gas such as hydrogen or nitrogen. The solution was cooled to about 5°C. and 152 g. (2 mol. with small excess) of n-butyraldehyde were added. While cooling and stirring vigorously, 100 g. of activated aluminium were added gradually. After addition of the activated aluminium, the reaction mixture was stirred rapidly for from two to four hours at about 25°C. The reaction mixture was then made just acid with sulfuric acid. The resulting solution was concentrated to a volume of about 2000 cc. and decolorized with activated charcoal. Upon cooling the decolorized solution N-(n-primarybutyl)-p-aminophenol sulfate separated out.

Upon dissolving the sulfate in water and making the solution just alkaline with sodium carbonate mixed with a little sodium sulfite, N-(n-primarybutyl)-p-aminophenol separated out in the form of fine white crystals melting at 70° to 71°C. Its nitroso derivative melted at 87° to 89°C.

The formula for N-(n-primarybutyl)-p-aminophenol is:



It can be converted into its salt, such as hydrochloride, by treating with aqueous solutions of the corresponding acids. I have found this substance to be a photographic developer and useful for inhibiting or retarding the deterioration of motor fuels.

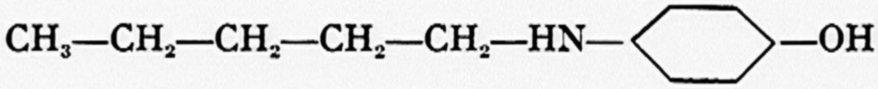
Example 3. *N-(n-primaryamyl)-p-aminophenol*

218 g. (2 mol.) of p-aminophenol were dissolved in 5000 cc. of water containing 142 g. (3.5 mol.) of sodium hydroxide. The solution was placed in a vessel equipped with a means of stirring and the air in the vessel displaced with a non-oxidizing gas, such as hydrogen or nitrogen. The solution was cooled to about 5°C. and 173 g. (2 mol.) of n-valeraldehyde were added. While cooling and stirring vigorously,

100 g. of activated aluminium were added gradually. After addition of the activated aluminium, the reaction mixture was stirred rapidly for from two to four hours at about 25°C. The reaction mixture was then made just acid with sulfuric acid. The resulting solution was concentrated to a volume of about 2500. and decolorized with activated charcoal. Upon cooling the decolorized solution, N-(n-primaryamyl)-p-aminophenol separated out.

Upon dissolving the sulfate in water and making the solution just alkaline with sodium carbonate mixed with a little sodium sulfite, N-(n-primaryamyl)-p-aminophenol separated out in the form of white crystals melting at about 73°C. Its nitroso derivative was an oil which did not solidify.

The formula for N-(n-primaryamyl)-p-aminophenol is:

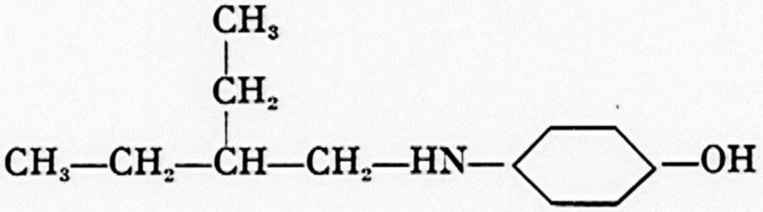


I have found this substance to be a photographic developer. This substance is also useful for retarding the deterioration of motor fuels.

Example 4. *N-(2-ethyl-n-primary butyl)-p-aminophenol*

218 g. (2 mol.) of p-aminophenol were dissolved in 5000 cc. of water containing 142 g. (3.5 mol.) of sodium hydroxide. The solution was placed in a vessel equipped with a means of stirring and the air in the vessel displaced with a non-oxidizing gas, such as hydrogen or nitrogen. The solution was cooled to about 50°C. and 200 g. (2 mol.) of 2-ethylbutanal were added. While cooling and stirring vigorously, 100 g. of activated aluminium were added gradually. After addition of the activated aluminium, the reaction mixture was stirred rapidly from two to four hours at about 25°C. The reaction mixture was then made just acid with sulfuric acid. The resulting solution was concentrated to a volume of about 2500cc. and decolorized with activated charcoal. Upon cooling, the decolorized solution N-(2-ethyl-n-primarybutyl)-p-aminophenol sulfate separated out.

Upon dissolving the sulfate in water and making the solution just alkaline with sodium carbonate mixed with a little sodium sulfite, N-(2-ethyl-n-primarybutyl)-p-aminophenol separated out in the form of white crystals melting at 98° - 99°. Its nitroso derivative was an oil which did not solidify. The formula of N-(2-ethylbutyl)-p-aminophenol is:



This new substance is an excellent motor fuel stabilizer serving to inhibit or retard deterioration thereof, I have found. In a similar manner N-(2-methyl-n-primarybutyl)-p-aminophenol can be prepared from 2-methylbutanal and p-aminophenol. This 2-methyl compound is likewise a good motor fuel stabilizer.

In a manner similar to that illustrated in the above examples aldehydes of two or more carbon atoms can be reacted with other aminophenols, e. g., m-aminophenol, 1-amino-2-methyl-4-hydroxybenzene, 1-amino-3, 4-dihydroxy benzene or the like. Benzaldehyde, salicylaldehyde or the like can be reacted with aminophenols as illustrated.

The strong base employed in my process is advantageously an alkali, such as sodium or potassium hydroxide. Lithium hydroxide can also be used. Quaternary ammonium hydroxides, such as benzyl-trimethyl ammonium hydroxide, can likewise be used. Aqueous strong bases are particularly useful, though methyl alcoholic or other solutions of strong bases can be employed.

The aldehyde and aminophenol are ordinarily employed in equimolecular proportions, though an excess of aldehyde can be employed. The strong base should be employed in quantity sufficient to dissolve the aminophenol and to react with the aluminium to generate sufficient hydrogen to form the N-substituted aminophenol.

The starting temperature is advantageously less than 10°C., the range 0° to 10°C. being suitable. During the process the reaction mixture can advantageously be held below 10°C., being allowed to rise slowly at the end of the process to around 25°C. ordinarily, although it can be held below 10°C. throughout. The temperature can be allowed to rise above 25°C. though this is generally less satisfactory.

It is advantageous to carry out my process in the absence of oxidants, such as oxidizing gases, e. g., air. However, it is not essential to displace the air in the reaction vessel, owing to the hydrogen which escapes during the process. The sodium sulfite employed in the above examples is to offset oxidation by air while precipitating the N-substituted aminophenols. The sodium sulfite can be replaced by sodium hydrosulfite or by corresponding potassium salts.

The aluminium reacts with the strong base generating hydrogen and dissolves in the excess strong base. Upon acidification with sulfuric acid the aluminium remains in solution as alum while the N-substituted aminophenol sulfate separates out. The aluminium is advantageously in a finely divided or activated form, in order to assure rapid reaction. The aluminum is advantageously added gradually to the reaction mixture. A convenient method of preparing an activated aluminium is as follows: 100 g. of aluminium in the form of small flakes, were treated in 10 g. portions with a solution of mercuric chloride, i. e., the 10 g. portion was added to 30 to 40 cc. of water containing 0.17 g. of mercuric chloride with stirring while cooling. The stirring was continued for about 10 minutes while the temperature rose to about 25° to 30°C. The so-prepared aqueous mixture was then employed, without separation of the activated aluminium, to add to the reaction mixture containing aminophenol, aldehyde and strong base.

The N-substituted aminophenols prepared by my process separate from the reaction mixture in a pure form. They can be recrystallized if desired from suitable solvents.

* * * *

What I claim as my invention and desire to be secured by Letters Patent of the United States is:

1. A compound selected from the group consisting of N-(n-primarybutyl)-aminophenols and N-(2-alkyl-n-primarybutyl)-aminophenols.

2. A compound selected from the group consisting of N-(n-primarybutyl)-p-aminophenols and N-(2-alkyl-n-primarybutyl)-p-aminophenols.

3. A N-(n-primarybutyl)-p-aminophenol.

4. N-(n-primarybutyl)-p-aminophenol.

5. N-(2-ethyl-n-primarybutyl)-p-aminophenol.

6. A salt selected from the group consisting of salts of N-(n-primarybutyl)-aminophenols and salts of N-(2-alkyl-n-primarybutyl)-aminophenols.

7. A process comprising reacting, in the presence of a strong base and aluminium, an aldehyde containing at least two carbon atoms with an aminophenol.

8. A process comprising reacting, in the presence of an aqueous alkali and activated aluminium, an aldehyde containing at least two carbon atoms with an aminophenol at from about 0° to about 10°C.

9. A process comprising reacting with alkali and activated aluminium a compound of the following formula:

$$R—CH{=}N—\text{phenylene}—OH$$

wherein R represents an alkyl group of at least two carbon atoms.

10. A process comprising reacting with aqueous alkali and activated aluminium, at from about 0° to about 10°C., a compound of the following formula:

$$R—CH{=}N\text{-phenylene}—OH$$

wherein R represents an alkyl group of at least two carbon atoms.

11. A process comprising reacting, in the presence of aqueous alkali and activated aluminium, n-butyraldehyde with p-aminophenol.

12. A process comprising reacting, in the presence of aqueous alkali and activated aluminium, n-valeraldehyde with p-aminophenol.

13. A process comprising reacting, in the presence of aqueous alkali and activated aluminium, a 2-alkylbutanal with p-aminophenol.

14. A process comprising reacting, in the presence of aqueous alkali and activated aluminium, 2-ethybutanal with p-aminophenol.

Pltf. Ex. No. 6

ELON LABORATORY

May 10, 1935.

To Mr. A. W. M. Dickins, Bldg. 46:—

RE:—N-ETHYL-p-AMINOPHENOL, N-BUTYL-p-AMINOPHENOL, and N-FURFURYL-p-AMINOPHENOL

I am sending you samples of N-ethyl-p-aminophenol, N-butyl-p-aminophenol, and N-furfuryl-p-aminophenol in the form of their sulfates.

Each compound has been tested for its behavior as a photographic developer. The preparation of the sulfates and the photographic tests were only incidental, since I was primarily interested in getting gasoline and oil soluble derivatives of p-aminophenol. The photographic tests show that the bases have reduction potentials that are relatively high (about the same as N-methyl-p-aminophenol). Since the bases are quite soluble in gasoline and oils but quite insoluble in water they give promise of being practical and potent antioxidants for gasoline, oils, etc. I shall also prepare the free bases and send you samples of them as soon as possible.

N-ETHYL-p-AMINOPHENOL

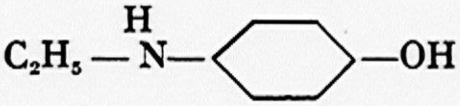
N-Ethyl-p-aminophenol was prepared by the reduction of ethylidene-p-aminophenol with activated aluminum in a solution of sodium hydroxide.

N-Ethyl-p-aminophenol sulfate was tried as a photographic developer. As is to be expected the free base is less soluble than N-methyl-p-aminophenol in sodium sulfite and sodium carbonate solutions but its lower solubility did not seriously interfere with its use as a photographic developer. The attached print was developed using the following formula:—

N-Ethyl-p-aminophenol sulfate	5.0	gms.
Sodium Sulfite	75.0	gms.
Sodium Carbonate	25.0	gms.
Potassium Bromide	2.5	gms.
Water	1	Liter

When sodium carbonate or sodium sulfite is added to a solution of N-ethyl-p-aminophenol sulfate the free base precipitates to form white crystals. Nitroso-ethyl-p-aminophenol is precipitated when a solution of sodium nitrite is added to an aqueous solution sulfuric acid solution of N-ethyl-p-aminophenol. N-Ethyl-p-aminophenol is soluble in dilute alkalis, dilute acids, and most organic liquids. 2 parts N-ethyl-p-aminophenol dissolved easily in 1000 parts of gasoline. The base is much less soluble in water than Elon base. The chemical structure of N-ethyl-p-aminophenol and its behavior as a photographic developer indicate that the compound has a relatively high reduction potential. These properties along with its solubility in most organic liquids (for example: esters, hydrocarbons) should make it an excellent antioxidant for gasoline and oils.

N-Ethyl-p-aminophenol melts at 111° - 112° C. Nitroso-ethyl-p-aminophenol melts at 81° - 83° C. with decomposition. The method of synthesis, the fact that it forms a nitroso derivative with nitrous acid, and its solubility in acids and alkalis indicate that the compound has the following structure:—



N-Ethyl-p-aminophenol was first prepared by Agfa in 1907, D.R.P. 205,415. They report the melting point of the base as 100° C. I found that the compound melted at 111° - 112° C. Agfa states that the nitroso derivative is a liquid. I found that the nitroso derivative was a solid which melted at 81° - 83° C. with decomposition.

N-BUTYL-p-AMINOPHENOL

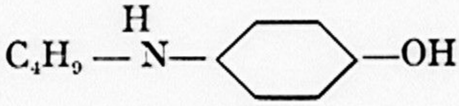
N-Butyl-p-aminophenol was prepared by the reduction of butylidene-p-aminophenol with activated aluminum in a solution of sodium hydroxide.

N-Butyl-p-aminophenol was tried as a photographic developer although difficulties arose because of its low solubility in sodium carbonate and sodium sulfite solutions. It closely resembles Elon in its behavior as a developer. The attached print was developed using the following formula:

N-Butyl-p-aminophenol sulfate	5.0	gms.
Sodium Sulfite	75.0	gms.
Sodium Carbonate	25.0	gms.
Potassium Bromide	2.5	gms.
Water	1	Liter

When sodium sulfite or sodium carbonate is added to a solution of N-butyl-p-aminophenol sulfate the free base N-butyl-p-aminophenol is precipitated as white crystals at once. Nitroso-butyl-p-aminophenol is precipitated when a solution of sodium nitrite is added to an aqueous sulfuric acid solution of N-butyl-p-aminophenol. N-Butyl-p-aminophenol is soluble in dilute alkalis, dilute acids, and most organic liquids. 2 Parts N-butyl-p-aminophenol dissolved easily in 1,000 parts of gasoline. The base is quite insoluble in water. The chemical structure of N-butyl-p-aminophenol and its behavior as an photographic developer indicate that the compound has a relatively high reduction potential. These properties along with its solubility in gasoline should make it an excellent antioxidant for gasoline and oils.

Pure N-Butyl-p-aminophenol melts at 70° - 71° C. Nitrosobutyl-p-aminophenol melts at 87° - 89° C. with decomposition. The method of synthesis, the fact that it forms a nitroso derivative with nitrous acid, and its solubility in acids and alkalis indicate that the compound has the following structure:



The compound is not referred to in the chemical literature and apparently has not been prepared before.

N-FURFURYL-p-AMINOPHENOL

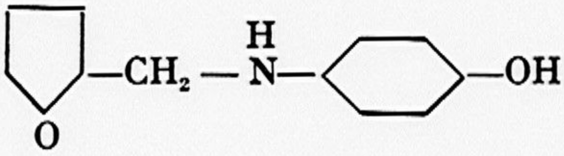
N-Furfuryl-p-aminophenol was prepared by the reduction of furfurylidene-p-aminophenol with activated aluminium in a solution of sodium hydroxide.

Attempts to use furfuryl-p-aminophenol sulfate as a photographic developer were unsuccessful because of the extremely low solubility of furfuryl-p-aminophenol base in sodium carbonate and sodium sulfite solutions.

A solution of N-furfuryl-p-aminophenol in aqueous sulfuric reacts with a solution of sodium nitrite to precipitate nitrosofurfuryl-p-aminophenol. When a solution of sodium carbonate or sodium sulfite is added to a solution of N-furfuryl-p-aminophenol sulfate the free base is precipitated at once as white crystals. The base is soluble in dilute alkalis, dilute acids, and most organic liquids. It is quite soluble in gasoline —over 2 parts in 1000, but insoluble in water. Its chemical structure indicates that its reduction potential is near that of Elon base, consequently it should be an excellent antioxidant for gasoline and oils, etc.

N-Furfuryl-p-aminophenol melts at 110° - 111°C. Its nitroso derivative melts at 95° - 98°C with decomposition.

The method of synthesis, the fact that it forms a nitroso derivative with nitrous acid, and its solubility in acids and alkalis indicate that the compound has the following structure:



The compound is not referred to in the chemical literature and apparently has not been prepared before.

F. R. Bean,
Elon Laboratory.

Pltf. Ex. No. 7

ELON LABORATORY

Oct. 5, 1936

Mr. D. I. Mayne, Patent Department.

Re: N-Furfuryl-p-aminophenol and
N-Butyl-p-aminophenol.

The accompanying bottles contains N-Furfuryl-p-aminophenol sulfate and N-Butyl-p-aminophenol sulfate. The free bases may be obtained therefrom by mixing each with hot water and then making alkaline to brilliant yellow paper with a mixture of sodium carbonate and sodium sulfite. On cooling the free bases crystallize and may be filtered off.

F. R. Bean

Pltf. Ex. No: 8

EASTMAN KODAK COMPANY
ROCHESTER, N. Y.

October 2, 1936

cc—Mr. H. G. Stone
Mr. H. Von Bramer
TE-4 Patent Application file
10-5-36

Mr. James C. White
Tenn. Eastman Corp.
Kingsport, Tenn.

Dear Mr. White:

I have asked Mr. Dickins to supply five gram samples of furfuryl, ethyl, propyl, butyl and amyl-aminophenols. These will be normal compounds and will be in the form of the sulphate. Dr. Bean has the furfuryl, ethyl and butyl compounds on hand and is preparing the propyl and amyl compounds. They are in the sulphate form because more stable and can easily be hydrolyzed back to the base in the presence of aqueous alkali. These samples will go forward to you as soon as I receive them.

My thought in sending these along is that it may expedite your determination of the inhibiting properties of these compounds for gasoline.

Yours very truly,

D. I. Mayne
Patent Department

DIM:BW

Pltf. Ex. No. 9

EASTMAN KODAK COMPANY
ROCHESTER, N. Y.

October 6, 1936

Mr. James C. White
Tenn. Eastman Corp.
Kingsport, Tenn.

Dear Mr. White:

We are sending you under separate cover bottles of the following samples:

5 grams N-Furfuryl-p-aminophenol sulphate
5 grams N-Ethyl-p-aminophenol sulphate
10 grams N-Butyl-p-aminophenol sulphate.

In transmitting these to me, Dr. Bean states that—

> "The free bases may be obtained therefrom by mixing each with hot water and then making alkaline to brilliant yellow paper with a mixture of sodium carbonate and sodium sulfite. On cooling the free bases crystallize and may be filtered off."

Yours very truly,
(Signed) D. I. Mayne (BW)
Patent Department

BW

Cc: Mr. H. G. Stone
Mr. Von Bramer
10/8/36

Pltf. Ex. No. 10

M. O. 973 Mr. Mayne

24266

Shop Order No.

Classification 23: 24.

SKETCH SHEET

Docket No. 5876.

Pat. Dept. File No.

EASTMAN KODAK COMPANY

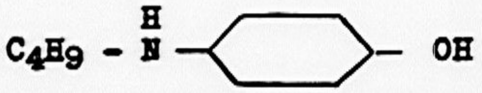
PATENT DEPARTMENT

INVENTOR: Fred R. Bean EMPLOYED AT: KODAK PARK

IMPROVEMENT IN: N-Butyl-p-Aminophenol Preparation of a New Compound, N-Butyl-p-aminophenol, and Use thereof as an Antioxidant and Inhibitor

N-Butyl-p-aminophenol, a derivative of p-aminophenol has not been prepared heretofore and has not been referred to in the chemical literature. It has now been prepared by the reduction of butylidene-p-aminophenol with dilute alkali and activated aluminum.

The free base, N-butyl-p-aminophenol, is a white crystalline solid melting at 70° - 71°C. Its nitroso derivative melts at 87° - 89°C. with decomposition. N-Butyl-p-aminophenol is soluble in dilute acids, dilute alkalis, and most organic liquids. The method of synthesis, the fact that it forms a nitroso derivative with nitrous acid, and its solubility in alkalis and acids indicate that the compound has the following structure:



From its chemical constitution and solubility in organic liquids I claim the compound as an antioxidant in the stabilization of gasoline, kerosene and petroleum distillates. I claim N-butyl-p-aminophenol also to be a useful antioxidant for preventing the deterioration of oils, fats, fatty oils, fatty esters and fatty acids, and as an inhibitor of oxidation and ageing of rubber, leather and analogous products.

EXAMPLE:

In a vessel provided with a stirrer 218 gms. p-aminophenol were dissolved in 5,000 cc. water containing 142 gms. sodium hydroxide. The solution was cooled to about 5°C. in an atmosphere of hydrogen or nitrogen and 186 cc. butyric aldehyde added. 100 gms. activated aluminum were then gradually added to the reaction mixture. After the reaction was complete the solution was acidulated with sulfuric acid, evaporated, and decolorized with charcoal. On cooling N-butyl-p-aminophenol sulfate crystallized out. On dissolving the N-butyl-p-aminophenol sulfate in water and making alkaline with sodium carbonate and a little sodium sulfite the base, N-butyl-p-aminophenol, separated out in the form of white crystals.

EASTMAN KODAK CO.
AUG 28 1936
PATENT DEPT.

IMPORTANT: Sign, Date and Witness: Forward to the Manager of the Works for the Patent Department.

Witnessed and Explained to me Aug 20 1936

Signatures of Witnesses Austin J. Gould

Leo J. Tanghe.

This sketch made by me on May 27, 1935

(Signature of person who made sketch.)

Fred R. Bean

(Signature of Inventor.)

(OVER)

EASTMAN KODAK COMPANY
PATENT DEPARTMENT

Shop Order No.
Classification

Docket No.
Patent Dept. File No.

INVENTION DATA SHEET

Application No.

May 27, 1935

Inventor **Fred R. Bean** Where Employed **KODAK PARK**

Improvement in N-Butyl-p-Aminophenol Preparation of a New Compound, N-Butyl-p-Aminophenol, and Use thereof as an Antioxidant and Inhibitor

Earliest Date of Invention March 12, 1934

How Can Date Be Proved Notebook

First Disclosed to Leo J. Vaughn On March 12 1934

First Sketches Made By On 19

Witnessed By

Model Designed By

Drawings Begun 19 Finished 19

Drawings No. Where Filed

First Model Made By

Model Begun 19 Finished 19

Model No. Where Filed

Model Photographed By On 19

Model Photograph No. Where Filed

Model Tested By On 19

Witnesses to Test

Report of Test

Remarks

Pltf. Ex. No. 12

TENNESSEE EASTMAN CORPORATION
KINGSPORT, TENNESSEE

October 26, 1936

Mr. D. I. Mayne
Eastman Kodak Company
343 State Street
Rochester, New York
R

Dear Mr. Mayne:

This will reply to your letter of October 20 in which you outline your plans for revision of our patent program on TE-2 and TE-4. Our people believe that your plans for submitting five additional patent applications as you have outlined them are quite satisfactory, and we think it is important to proceed with this program as quickly as possible.

We agree that it would probably be advisable to submit drafts of certain of these applications to Dr. Morrell as we have in the past.

I am enclosing copy of Mr. Von Bramer's Report No. 602-6 headed "Analysis of TE-2 and TE-4" which will give you the induction results on the N. amyl-p-amino phenol and the N. butyl-p-amino phenol. The small sample of furfuryl aminophenol which Dr. Bean sent us was insufficient for us to run complete tests, and we wonder if you will ask Dr. Bean to submit to us a larger quantity. We would need at least 50 grams of this material, in fact, it would be better if we could have as much as 100 grams. We shall send on to you our results of the induction tests of the furyfuryl compounds as well as the propyl and amyl compounds as soon as these materials have been received from Dr. Bean and we have been able to complete our tests.

Yours very truly,

James C. White James C. White

Pltf. Ex. No. 15

REPORT ON THE INVESTIGATIONS OF THE CLIFFORD U. S. PATENT 2,109,164

October 10, 1941

At the request of Mr. D. I. Mayne, attorney for the Eastman Kodak Company of Rochester, New York, the following investigations were carried out by Dr. Frederic R. Bean or by Mr. R. Stoddard Vaughn under the supervision of Dr. Frederic R. Bean.

Attempted Preparation of N-(n-primary butyl)-p-aminophenol

The preparation of N-(n-primary butyl)-p-aminophenol according to the process set forth by Clifford in U. S. patent 2,109,164, viz. "by reacting butyl amine and p-chlorophenol in an autoclave", was attempted. No N-(n-primary butyl)-p-aminophenol was obtained when n-primary butyl amine and p-chlorophenol were heated in an autoclave.

Attempted Preparation of N-isobutyl-p-aminophenol

The preparation of N-isobutyl-p-aminophenol according to the process set forth by Clifford in U. S. patent 2,109,164, viz. "by reacting butyl amine and p-chlorophenol in an autoclave", was attempted. No N-isobutyl-p-aminophenol was obtained when isobutyl amine and p-chlorophenol were heated in an autoclave.

The actual data pertaining to the above investigations are summarized below:

> 257 g. (2 mol.) of p-chlorophenol and 146 g. (2 mol.) of n-primarybutylamine were heated in a steel autoclave for about four hours at 200° to 220°C. After the reaction mixture had cooled, it was made strongly acid with concentrated hydrochloric acid. The resulting acidified mixture was diluted to a volume of about three liters. Unreacted p-chlorophenol separated as an oil.

The chlorophenol was removed in a separatory funnel. The aqueous portion was treated with sodium chloride to salt out further chlorophenol. The salted solution was extracted twice with separate 250 cc. portions of diethyl ether to remove the remainder of the chlorophenol. The remaining aqueous liquors were boiled to remove traces of chlorophenol and ether. The so-treated aqueous liquors were made strongly alkaline with sodium hydroxide and then boiled until all unreacted butylamine was volatilized. The resulting aqueous liquors were made acid with hydrochloric acid. A portion of the acid liquor was treated with a freshly prepared concentrated aqueous solution of sodium nitrite. No solid precipitate nor oil formed, showing the absence of N-(n-primarybutyl)-p-aminophenol.

130 grams (1 mol.) of para chlorophenol and 75 grams (1 mol.) of isobutyl amine were heated in a steel autoclave for four hours at 190° - 195°C. One and one-half hours were required for the autoclave temperature to attain 190°C. and it was cooled overnight. The reaction mixture was removed from the autoclave and diluted with two liters of water. This mixture was then made definitely acid with concentrated hydrochloric acid and steam distilled to remove the portion of unreacted p-chlorophenol. The remaining p-chlorophenol was extracted with two 250 cc. portions of ether. The remaining aqueous liquors were made definitely alkaline with sodium hydroxide and boiled to remove unreacted iso butyl amine. The resulting liquors were acidified with hydrochloric acid. A portion of the acid liquor was treated with a freshly prepared concentrated aqueous solution of sodium nitrite. No solid precipitate nor oil formed, showing the absence of N-isobutyl-p-aminophenol.

Frederic R. Bean
R. Stoddard Vaughn

Pltf. Ex. No. 21

REPORT ON INVESTIGATIONS OF CLIFFORD U. S. PATENT 2,109,164

October 10, 1941

At the request of Mr. D. I. Mayne, attorney in the Patent Department of the Eastman Kodak Company, Rochester, New York, the following investigations were carried out by Dr. Lee G. Davy:

Attempted Preparation of N-(n-primary butyl)-p-aminophenol

The preparation of N-(n-primary butyl)-p-aminophenol according to the process set forth by Clifford, viz. "by reacting butyl amine and p-chlorophenol in an autoclave", was attempted. No N-(n-primary butyl)-p-aminophenol was obtained when n-primary butyl amine and p-chlorophenol were heated in an autoclave.

Attempted Preparation of N-isobutyl-p-aminophenol

The preparation of N-isobutyl-p-aminophenol according to the process set forth by Clifford, viz. "by reacting butyl amine and p-chlorophenol in an autoclave", was attempted. No N-isobutyl-p-aminophenol was obtained when isobutyl amine and p-chlorophenol were heated in an autoclave.

The actual data pertaining to the above investigations are summarized below:

> 219 grams (3 moles) of normal primary butyl amine and 128.5 grams (1 mole) of p-chlorophenol were heated together in a stainless steel autoclave at 210°C for 18 hours. The autoclave was cooled and opened. The contents of the autoclave were placed in a one liter round bottom flask. To the contents were added 48 grams of sodium hydroxide dissolved in 200 cc. of water. The resulting mixture was distilled to remove the n-primary butyl amine. The residue remaining after the distillation was made strongly acid with 172 grams of concentrated hydrochloric acid dissolved in 172 grams of water. The resulting mixture was then extracted three times with 200 cc. portion of diethyl ether to remove unused p-chlorophenol. To a 5 cc. portion of the remaining acidic aqueous solution was then added 1 cc. of a 10 per cent (by weight) aqueous solution of sodium nitrite. No solid precipitate nor oil was formed, showing that no N-(n-primary butyl)-p-aminophenol had been formed.
>
> 221 grams (3.02 moles) of isobutyl amine and 128.5 grams (1 mole) of p-chlorophenol were placed in a stain-

less steel autoclave. The autoclave was closed and then heated at 200°C for eight hours. The autoclave was cooled and opened. The contents of the autoclave were placed in a one liter round bottom flask. To the contents were added 48 grams of sodium hydroxide dissolved in 200 cc. of water. The resulting mixture was distilled to remove the unused isobutyl amine. The residue remaining after the distillation was made strongly acid with 172 grams of hydrochloric acid dissolved in 172 grams of water. The resulting mixture was then extracted five times with 100 cc. portions of diethyl ether to remove unused p-chlorophenol. To a 5 cc. portion of the remaining acidic aqueous solution was then added 1 cc. of a 10 per cent (by weight) aqueous solution of sodium nitrite. No solid precipitate nor oil was formed, showing that no N-isobutyl-p-aminophenol had been formed.

Lee G. Davy

Pltf. Ex. No. 25

SPECIFICATIONS
of
UNITED STATES PATENT APPLICATION
of
Thomas H. Rogers and Vanderveer Voorhees
of
Whiting, Indiana
for:
"MOTOR FUEL PRODUCTS"

Serial No. 427416 filed February 10, 1930

The present invention relates to improvements in hydrocarbon motor fuel products of the character of gasoline, and has to do more particularly with motor fuel products derived by the cracking of higher boiling hydrocarbon oils of such types as display a tendency to form gum or resinification products on storage, even after refining treatment with a dilute sulfuric acid such as sulfuric acid of 75% strength or lower. This tendency may be indicated by an accelerated test designated hereinafter as the oxygen absorption test, which is a measure of the potential gum formation or resinification properties of the motor fuel products.

In methods of cracking high boiling hydrocarbon oils to produce lower boiling hydrocarbon oils, the products which have been secured may in general be grouped in two classes; the first, which at the present time forms the bulk of pressure cracked gasoline, having an anti-knock value somewhat greater but not substantially greater than straight-run gasoline; a somewhat lower Baumé gravity than straight-run gasoline; containing a moderate proportion of unsaturated compounds as determined by combined absorption and polymerization by sulfuric acid in accordance with the method of determination hereafter set forth; and being in general of such a character that the constituents thereof which tend to form gum on storage and handling are readily removed by treatment with sulfuric acid to produce a product comparable in that respect to straight-run gasoline. Motor fuel products of the second general class, hitherto derived principally from vapor phase processes or very high temperature pressure cracking processes, have anti-knock values substantially higher than straight-run gasoline; have a much lower A. P. I. gravity; contain large amounts of unsaturateds as determined by sulfuric acid absorption and polymerization; and require excessive treatment with sulfuric acid, with very substantial losses, to remove potential gum-forming constituents.

Although it cannot be said that there is a hard and fast line between the two types of products, the former type of product, with 425° F. end point, will in general have an anti-knock value not exceeding an equivalent of 1.2 cubic centimeters of tetra-ethyl lead per gallon of straight-run Mid-continent gasoline; an olefin content as determined by sulfuric acid absorption and polymerization of not over 40%; an A. P. I. gravity of 56 to 59°; and an oxygen absorption induction period greater than 125 minutes, as determined by the test hereafter set forth. The second type of product, which may be designated as the vapor phase type, will in general have an anti-knock value equivalent to at least 1.2 cc. tetraethyl lead per gallon straight-run Mid-Continent gasoline, an A. P. I. gravity of about 49 to 55°; an unsaturated content as determined by sulfuric acid absorption and polymerization exceeding 40%, and usually above 45%; and an oxygen absorption induction period of less than 125 minutes.

The latter type of product in general tends to form gum, even on storage in the dark, and under like conditions, tends to lose its anti-knock properties. The same tendencies may exist, to a greater or less extent, in products of the first mentioned type and, the present invention may be employed in connection therewith, although particularly intended for use in connection with products of the so-called vapor-phase cracked type.

Thus, products of certain processes commonly designated as liquid phase or liquid-vapor phase cracked products, although having a lower unsaturated content than vapor phase cracked products, as shown by combined absorption and polymerization by sulfuric acid, have gum forming properties similar to those of vapor phase cracked products. The present invention may be applied in connection with such products, as well as with such highly cracked products as are derived by drastic or high temperature cracking or destructive distillation of bituminous materials, such as coal, peat, lignite, and the like.

In determining the oxygen absorption induction period, by which an indication of the potential gum-forming properties of the gasoline on storage and handling may be secured, the following procedure may be employed; 100 cubic centimeters of the motor fuel or gasoline to be tested are placed in a one liter flask, the total volumetric capacity of which is 1150 to 1200 cubic centimeters. The gasoline is maintained at about 212° F. by heating in a steam bath while an oxygen atmosphere is maintained in the flask under a substantially constant pressure of about 2½ atmosphere. The flask is vigorously agitated and the volume of oxygen absorbed is measured at frequent intervals. The oxygen absorption is characterized by an initial period in which relatively small amounts are absorbed and subsequent to this period there is a rapid increase in the rate of absorption. This initial period of slow absorption is designated the induction period, and its end is regarded as the point at which oxygen absorption under the conditions stated reaches a rate of 1 cubic centimeter per minute. If the oxygen absorption is plotted against the time in minutes with equal distances on the coordinates for time in minutes and absorption in cubic centimeters, it will be evident that the end of the induction period will be indicated upon the curve by the point where the tangent to the curve has a slope of 45°. The length of the induction period, as determined by this test, provides a fairly accurate indication of the potential gum-forming behavior of the motor fuel product on storage and handling.

The motor fuel products with which the present invention is primarily concerned, are produced from cracked distillate products of the vapor-phase cracked type which, after treatment with dilute sulfuric acid of 50 to 70% strength, have an oxygen absorption induction period of less than 125 minutes. The following procedure has been employed in producing such a product.

A hydrocarbon oil, preferably a distillate product heavier than gasoline, is forced through a continuous coil in a heating zone and brought therein to an outlet temperature of 825 to 975° F. while maintaining thereupon a pressure exceeding 200 lbs. under conditions to produce from 15 to 35% of hydrocarbons in the gasoline boiling point range in the products leaving the coil. The material leaving the coil is preferably reduced in pressure and fractionally condensed to separate the hydrocarbons in the gasoline range of boiling points as a distillate. The vaporization is controlled, or the distillate products may be rerun, to produce a material conforming to the desired specifications, for example, to 425° F. end point.

Such a material has a gravity of 48 to 56° Baumé as compared with 58 to 60° Baumé for a straight-run gasoline of similar boiling range from Mid-Continent crude. The product, without treating, is found to have an oxygen absorption induction period of about 35 to 70 minutes. It is suitably treated with dilute sulfuric acid, say 60% acid, to the extent of 6 lbs. of such acid per barrel of distillate. The loss on treating is about ½%, the color of the product is not substantially altered, but is substantially stabilized and the oxygen absorption induction period is increased to 80 to 90 minutes. In determining the unsaturateds in such a product it shows a combined absorption and polymerization loss of 50% to 70% when the test is made with 90% sulfuric acid. This loss is determined by subjecting a given volume of the material to treatment at room temperature with two volumes of 90% sulfuric acid, separating the remaining oil from the acid layer and redistilling the unabsorbed oil to produce a distillate having the same end point as the original material treated. The loss in volume relative to the original material is that which is designated as the combined absorption and polymerization loss.

A treated distillate product, produced as hereinbefore described, or other cracked products having a tendency to form gum or resinification products on storage, is treated by adding thereto a small proportion of an organic inhibiting agent or anti-oxidant having reducing properties and containing an amino-hydroxy aromatic radical, including the aminophenols and substituted aminophenols. Such substances are employed in proportions of, in general, less than 0.1% and preferably less than 0.05%, and substantially stabilize the product against gum formation and resinification, and also against loss in anti-knock. This stabilization is indicated by an increase in the oxygen absorption induction period of the products to which they are added. As examples of such materials, we employ the aminophenols, para- and ortho-methylaminophenol, dimethylaminophenol, hydroxyazobenzine, butyl, isopropyl and benzyl aminophenols, dihydroxy diphenyl amine, paraphenetidine, amino naphthol and the like.

The specific activity of these materials varies to a considerable extent, all of them, however, being effective in inhibiting the gum-forming properties of the motor fuel product and preventing its deterioration, for example, by decrease of its anti-knock value, and these properties are indicated by an increase in the induction period of the product. It is preferred to employ those which, when added to a vapor-phase type of product as hereinbefore set forth, in proportions of 0.05% or less, will increase the induction period to above 125 minutes and preferably to 300 minutes or more. Thus, with the product prepared as above described, and which, after dilute acid treatment has an induction period of 55 minutes and a lead equivalent of 1.7 cubic centimeters tetra-ethyl lead per gallon of straight-run Mid-Continent gasoline, the addition of 0.005% of ortho- or of para-aminophenol increases the induction period to about 275 minutes or to 440 minutes respectively. 0.01% of para-methylaminophenol increases the induction period to 1800 minutes and 0.001% to 435 minutes. 0.005% of para-phenetidine increases the induction period to above 125 minutes.

The results secured by the application of the oxygen absorption test, as hereinbefore described, definitely indicate the behavior of the motor fuel product, both in its tendency to form gum and to lose in anti-knock value. Thus, two lots of the same gasoline were stored for eight months, one without any added constituents and the other with 0.002% of para-methylaminophenol added. At the end of eight months, the untreated gasoline showed a gum test of 42 milligrams of non-volatile resin, whereas the lot treated with paramethylaminophenol showed the formation of only 2 milligrams. Similarly, samples of gasoline produced by cracking Mid-Continent gas oil in vapor phase and having an anti-knock value equivalent to 1.7 cubic centimeters of tetra-ethyl lead per gallon of Mid-Continent gasoline, without added constituents, at the end of seven months, had an anti-knock equivalent of zero; that is, no higher than Mid-Continent gasoline. The same product, to which 0.002% of para-methylaminophenol was added, and which was stored under similar conditions for the same period of time, showed at the end of that period an anti-knock value of 1.5 cubic centimeters or only slightly under the original anti-knock value had when placed in storage. These results are typical of those secured by the organic reducing agents containing the aminohydroxy aromatic radical, added in accordance with the present invention as hereinbefore set forth.

The marked extension of the oxygen absorption induction period by the addition of the substances having the characteristics of the bodies hereinbefore described is not accompanied by any change in the anti-knock value, 90% acid absorption and polymerization loss or other characteristic property of the motor fuel.

Motor fuels produced in accordance with the present invention may be recognized by the fact that their oxygen absorption induction period is decreased by successive treatments with dilute acid and dilute alkali. In the preferred form of the invention, in which the motor fuel product shows a loss of 40% or higher by absorption and polymerization by 90% sulfuric acid, this decrease may be from upwards of 125 minutes, and generally upwards of 300 minutes, to less than 100 minutes. Thus a vapor-phase pressure distillate product having an anti-knock value equivalent to about 2 cubic centimeters of tetra-ethyl lead per gallon of straight-run gasoline and showing a loss on absorption and polymerization by 90% sulfuric acid exceeding 50%, had an induction period of approximately 45 minutes. After treatment with 6 lbs. per barrel of 60% sulfuric acid, the anti-knock value and sulfuric acid absorption and polymerization loss were substantially unchanged and the induction period increased to approximately 86 minutes. The addition of 0.002% of para-methylaminophenol increased the induction period to about 520 minutes without affecting the other characteristics of the motor fuel. This product constitutes a preferred embodiment of the present invention. On washing a given volume of it successively with 10% by volume of 50% sulfuric acid or other dilute sulfuric acid of insufficient strength to effect treating, and 10% by volume of approximately 10% alcoholic sodium hydroxide solution, the oxygen absorption characteristics were altered, the induction period being reduced to about 58 minutes.

If desired, the anti-knock value of the motor fuel described herein may be increased by the addition of small amounts of tetra-ethyl lead or other suitable material and it is intended that the accompanying claims shall include the products of the type herein set forth regardless of the addition thereto of tetra-ethyl lead or other materials modifying their fuel characteristics.

This application is in part a division and continuation of our prior application Serial No. 211,554, filed August 8, 1927.

We claim:

1. The method of preserving cracked hydrocarbon distillates which deteriorate and develop gums on storage, by incorporating therein a small proportion of an organic reducing agent containing an aminohydroxy aromatic radical.

2. The method of preserving cracked hydrocarbon distillates which deteriorate and develop gums on storage, by incorporating therein a small proportion of an organic reducing agent containing an aminophenol radical.

3. The method of preserving cracked hydrocarbon distillates which deteriorate and develop gums on storage, by incorporating therein a small proportion of an aminophenol.

4. The method of preserving cracked hydrocarbon distillates which deteriorate and develop gums on storage, by incorporating therein a small proportion of a substituted aminophenol.

5. The method of preserving cracked hydrocarbon distillates which deteriorate and develop gums on storage, by incorporating therein a small proportion of a substituted aminophenol in which the substituting group is substituted for an amino hydrogen.

6. The method of preserving cracked hydrocarbon distillates which deteriorate and develop gums on storage, by incorporating therein a small proportion of a methylaminophenol.

7. The method of preserving cracked hydrocarbon distillates which deteriorate and develop gums on storage by incorporating therein a small proportion of benzyl aminophenol.

8. The method of preserving a vapor-phase cracked hydrocarbon fuel product having an anti-knock value equivalent to upwards of 1.3 cubic centimeters tetra-ethyl lead per gallon straight-run Mid-Continent gasoline and showing a loss of upwards of 40% by absorption and polymerization by 90% sulfuric acid and having an oxygen absorption induction period of less than 125 minutes, by incorporating therein not more than 0.1% of an organic reducing agent containing an aminohydroxy aromatic radical.

9. The method of preserving a vapor-phase cracked hydrocarbon motor fuel having an anti-knock value equivalent to upwards of 1.3 cubic centimeters of tetra-ethyl lead per gallon of straight-run Mid-Continent gasoline, and showing a loss of upward of 40% by absorption and polymerization by 90% sulfuric acid by incorporating therein not more than 0.1% of an organic reducing agent containing an aminophenol radical whereby the oxygen absorption induction period is increased to upward of 125 minutes.

10. The method of preserving a vapor-phase cracked hydrocarbon distillate motor fuel having an anti-knock value equivalent to upwards of 1.3 cubic centimeters of tetra-ethyl lead per gallon of straight-run Mid-Continent gasoline and showing a loss of upward of 40% by absorption and polymerization by 90% sulfuric acid by incorporating therein not more than 0.05% of a substituted aminophenol.

11. The method of preserving a vapor phase cracked hydrocarbon distillate motor fuel having an anti-knock value equivalent to upward of 1.3 cubic centimeters of tetra-ethyl lead per gallon of straight-run Mid-Continent gasoline and

showing a loss of upward of 40% by absorption and polymerization by 90% sulfuric acid, by incorporating therein not more than 0.05% of para-methylaminophenol, whereby the oxygen absorption induction period of said fuel is increased to upward of 125 minutes.

12. A motor fuel product consisting substantially entirely of cracked hydrocarbon distillates tending to deteriorate and develop gums, said product containing a small proportion of an organic reducing agent having an aminohydroxy aromatic radical.

13. A motor fuel product consisting substantially entirely of cracked hydrocarbon distillates which tend to deteriorate and develop gums, said product containing a small proportion of an organic reducing agent having an aminophenol radical, whereby its tendencies to deteriorate and develop gums are inhibited.

14. A hydrocarbon motor fuel product consisting substantially entirely of cracked hydrocarbon distillates which tend to deteriorate and develop gums, said product containing a small proportion of an aminophenol, whereby its tendencies to deteriorate and develop gums are inhibited.

15. A motor fuel product consisting substantially entirely of cracked hydrocarbon distillates which tend to deteriorate and develop gums, said product containing a small proportion of a substituted aminophenol, whereby its tendencies to deteriorate and develop gums are inhibited.

16. A motor fuel product consisting substantially entirely of vapor-phase cracked hydrocarbon distillates and being of a character showing a loss of upward of 40% by absorption and polymerization by 90% sulfuric acid, said product containing a small proportion of an organic reducing agent having an aminohydroxy aromatic radical.

17. A motor fuel product consisting substantially entirely of vapor-phase cracked hydrocarbon distillates and being of a character showing a loss of upward of 40% by absorption and polymerization by 90% sulfuric acid, said product con-

taining a small proportion of an organic reducing agent having an aminophenol radical.

18. A motor fuel product consisting substantially entirely of vapor-phase cracked hydrocarbon distillates and being of a character showing a loss of upward of 40% by absorption and polymerization by 90% sulfuric acid, said product containing a small proportion of an aminophenol and having an oxygen absorption induction period of upward of 125 minutes.

19. A motor fuel product consisting substantially entirely of vapor-phase cracked hydrocarbon distillates and being of a character showing a loss of upward of 40% by absorption and polymerization by 90% sulfuric acid, said product containing a small proportion of a substituted aminophenol.

20. A motor fuel product consisting substantially entirely of vapor-phase cracked hydrocarbon distillates and being of a character showing a loss of upward of 40% by absorption and polymerization by 90% sulfuric acid, said product containing a small proportion of methylaminophenol.

21. A motor fuel product consisting substantially entirely of vapor-phase cracked hydrocarbon distillates and being of a character showing a loss of upward of 40% by absorption and polymerization by 90% sulfuric acid, said product containing a small proportion of benzyl aminophenol.

Evidence on Behalf of the Defendant

Mr. Moore. If there is no objection I will introduce a copy of the Patent Examiner's statement before the Board of Appeals as Defendant's Exhibit 1, * *.

The Court. There is no objection to that. It will be received as * * * Defendant's Exhibit No. 2 in Civil Action No. 12618.

(Document containing patents, statement of Examiner, etc., in Civil Action 12,618, was marked Defendant's Exhibit No. 2 and received in evidence.)

(Defendant's Exhibit No. 2 contains the following documents:)

D — Rogers et al., 2,084,977
E — Clifford, 2,109,164
F — British patent, 350,438
G — Statement of the Examiner,
H — Decision of the Board of Appeals,
I — Decision of the Board of Appeals on Request for Reconsideration.

* * * *

Mr. Moore. If your Honor please, I would like to have the Von Bramer patent used as the basis of my cross examination, marked for identification as Defendant's Exhibit 3.

(Von Bramer patent was marked Defendant's Exhibit 3 for identification.)

UNITED STATES PATENT OFFICE

2,084,977

MOTOR FUEL PRODUCTS

Thomas H. Rogers and Vanderveer Voorhees, Hammond, Ind., assignors, by mesne assignments, to Gasoline Antioxidant Company, Wilmington, Del., a corporation of Delaware

No Drawing. Application March 26, 1937, Serial No. 133,187

2 Claims. (Cl. 44—9)

This invenion relates to the stabilization of motor fuels and particularly to the stabilization of cracked gasolines which tend to deteriorate and develop gums.

In particular, the present invention relates to the treatment of highly cracked gasolines or hydrocarbon motor fuel distillates of the type which are normally unstable, particularly with respect to gum formation on storage, and which, having an initial anti-knock rating superior to that of straight-run gasoline, tend to deteriorate in anti-knock rating.

The tendency of products of this type to develop gum forming constituents, is indicated, for example, by their oxygen absorption induction period, determined, for example, as described in the article by Voorhees and Eisinger in Proceedings of the American Petroleum Institute, Section 2, January 3, 1929, page 170. The oxygen absorption induction period is the period of time for which the rate of absorption of oxygen by a 100 cc. sample of motor fuel maintained in a liter flask at an elevated temperature of 210–212° F. in an atmosphere of oxygen under about 1.5 to 2 atmospheres pressures remains below 1 cc. per minute.

It has been customary to treat motor fuels of the type indicated above with substances such as strong sulphuric acid, fuller's earth and the like to remove these gum-forming constituents. Such treatments remove a substantial amount of the unsaturated constituents in the gasoline. Since these unsaturated constituents are partly responsible for the high anti-knock properties of cracked gasoline, it is apparent that such treatment destroys some of the desirable qualities of cracked gasolines. In addition, these treatments result in a loss in volume of motor fuel.

The object of the present invention is to provide a new class of organic materials that may be added to these gum-forming cracked gasolines, in small amounts, to inhibit or substantially retard the formation of gum therein. The amount of these gum inhibitors employed to stabilize a cracked gasoline will vary with the particular cracked gasoline to be treated but, in general, the amount of the inhibitor used will vary from 0.01 to 0.001%. With some motor fuels, the amount used will be as low as 0.0006%. Even in these small concentrations, the class of inhibitors herein disclosed have proven to be very effective as gum inhibitors in cracked gasoline.

Our present invention relates to the use of ortho-, meta-, and para-butyl aminphenols as gum inhibitors for cracked gasolines. In other words, these inhibitors are aminophenol wherein one amino hydrogen has been substituted by a butyl group, particularly the normal butyl radical. The corresponding secondary butyl, tertiary butyl and iso-butyl aminophenols may also be used. In addition, the benzene ring of the compound may be substituted with hydrocarbon radicals such as the methyl, ethyl and propyl radicals.

To illustrate the effectiveness of the above mentioned compounds, 0.002% of para-n-butyl aminophenol was added to a cracked gasoline having an initial induction period of 250 min. and it was found that this amount of the inhibitor raised the induction period to 875 minutes. In addition, these inhibitors do not develop objectionable color in the treated gasoline even upon exposure to sunlight.

In addition to using the compounds indicated above as gum inhibitors in cracked gasoline, these agents may be used in blends of motor fuels containing substantial amounts of gum-forming cracked gasoline. In using the above mentioned compounds to retard gum formation in cracked gasolines, we prefer to add the inhibitor to fresh gasoline or prior to the formation of gum.

This application is a continuation-in-part of our co-pending application Serial 427,416, filed February 10, 1930.

We claim:

1. A motor fuel product comprising cracked hydrocarbon distillates of the character of gasoline which normally tend to deteriorate and develop gum, said product containing a small proportion of aminophenol wherein one amino hydrogen has been substituted by a butyl radical, in quantities sufficient to retard such gum formation.

2. The method of preserving cracked hydrocarbon distillates of the character of gasoline which normally tend to develop gum on storage, which comprise incorporating therein a small proportion of aminophenol wherein one amino hydrogen has been substituted by a butyl radical.

THOMAS H. ROGERS.
VANDERVEER VOORHEES.

Patented Feb. 22, 1938 2,109,164

Deft. Ex. No. 2-E

UNITED STATES PATENT OFFICE

2,109,164

RUBBER COMPOSITION AND METHOD OF PRESERVING RUBBER

Albert M. Clifford, Cuyahoga Falls, Ohio, assignor to Wingfoot Corporation, Wilmington, Del., a corporation of Delaware

No Drawing. Application October 22, 1932, Serial No. 639,151

2 Claims. (Cl. 18—50)

This invention relates to the retarding of the ageing of deteriorable organic compositions; more particularly, to the use in rubber, balata, gutta percha, transformer oils, gasoline and other deteriorable organic compositions of age retarders or antioxidants of the kind hereinafter set forth. The substances to which the invention relates have been found by actual test to protect against deterioration from such influences as heat, light and oxygen. They have also been found in the case of rubber to materially enhance the resistance to deterioration by flexing. In general they accomplish various desirable ends, among them, in the case of rubber, that of imparting improved tensile strength to the treated product and, in the case of gasoline, of preventing the formation of gums and gummy materials.

Substances which may be employed as antioxidants in accordance with the teachings of the present invention are, or may be regarded as, primary, secondary, and tertiary amines of the type formula $(R_1).N.(R).X.(R_2)$ wherein (R) is an alkyl, aryl or aralkyl radical attached by either a single or double bond to N and by a single bond to X, such radical being preferably unsubstituted, but having, if desired, hydroxy, amino, nitro, halogen, alkoxy, aryloxy, aralkoxy or other substituents. Thus (R), where substituted, may be called an organic radical, and, where unsubstituted, a hydrocarbon group. In the formula, X represents oxygen, sulfur, selenium or tellurium, all of which are members of Group VI of the Periodic System, and N stands, as preferred, for a single nitrogen atom or for the nitrogen-containing group

$$\overset{|}{\underset{|}{N}}\text{—}N=O.$$

Groups (R_1) and (R_2) embrace radicals selected from a class consisting largely of alkyl, aryl, aralkyl; halogen alkyl, halogen aryl, halogen aralkyl; nitro alkyl, nitro aryl, nitro aralkyl; amino alkyl, amino aryl, amino aralkyl; hydroxy alkyl, hydroxy aryl, hydroxy aralkyl; alkoxy alkyl, alkoxy aryl, alkoxy aralkyl; aryloxy alkyl, aryloxy aryl, aryloxy aralkyl and like radicals and, in the case of (R_1), of hydrogen as well. Except where (R) is attached to N by two bonds and, further, where N represents the nitrogen-containing group

$$\overset{|}{\underset{|}{N}}\text{—}N=O$$

conditions under which Group (R_1), like Group (R_2), can embrace one and only one univalent radical, Group (R_1), may consist either of one bivalent radical or of two univalent radicals, the latter, where present, being either like or unlike each other. Group (R_2) may be a heterocyclic radical, differing in this respect from Group (R_1); but the latter, as is also true of (R), may represent a bivalent hydrocarbon chain constituting a part of a nitrogen-heterocyclic radical embodying N where the latter is a single nitrogen atom. Within these limits (R_1) and (R_2) may be either like or unlike; i. e., they may be composed, as the case may be, of radicals which are entirely like each other, which are partly like each other, or which are wholly unlike each other.

Examples of compounds falling within the type formula given above are dicyclohexyl amino chlorphenyl tolyl ether, wherein Group (R_1) includes the two univalent cyclohexyl radicals, Group (R_2) is made up of the tolyl nucleus, (R) is the chlorphenyl radical, and X is oxygen; p-ethoxy phenyl naphthyl nitrosamine, wherein N is the group

$$\overset{|}{\underset{|}{N}}\text{—}N=O$$

Group (R_1) is the naphthyl radical, Group (R_2) is the ethyl radical, X represents oxygen and (R) is the phenyl nucleus; p-amino tolyl di-ethyl ether, wherein Group (R_1) consists of two hydrogen atoms, X is oxygen, and (R) and Group (R_2) represent ethyl radicals; 1-methoxy 4-piperidyl benzene, wherein (R) is the benzene ring, X and (R_2) are respectively the oxygen and methyl radical of the methoxy group, N is the heterocyclic nitrogen atom, and (R_1) is the bivalent hydrocarbon chain which, with the heterocyclic nitrogen atom, makes up the piperidyl group; and p-methoxy pipecoline, wherein Group (R_1) consists of a single hydrogen atom, N is the heterocyclic nitrogen atom, (R) is the hydrocarbon part of the pipecoline nucleus, X stands for oxygen and Group (R_2) is the methyl radical of the methoxy substituent.

Falling within the scope of the invention are, for example, para phenoxy diphenyl amine, para para' dibutoxy diphenyl amine, N-tolyl phenetidine, 2-methyl 4-methoxy diphenylamine, 2-4-diamino 4' butoxy diphenyl amine, para hydroxy para' methoxy diphenyl amine, para hydroxy para' ethoxy diphenyl amine, 3-amino 4-methoxy diphenyl amine, 3-ethyl 4-methoxy diphenyl amine, 3-methoxy 4-hydroxy 4' hydroxy diphenyl amine, N-phenyl phenetidine, 3-methoxy 3' hydroxy diphenyl amine, 2-4-diamino 4-hydroxy diphenyl ether, 3-chlor 4-methoxy 4' hydroxy diphenyl amine, 3-chlor 4-butoxy diphenyl amine, 3-nitro 4-methoxy diphenyl amine, 3-ethoxy 4-hydroxy diphenyl amine, 3-ethoxy 4-hydroxy ditolyl amine, p-ethoxy phenyl anthracyl amine, methoxy phenyl tolyl amine, etc.

Others are 3′ methyl 4-phenoxy diphenyl amine, 3′ ethyl 4-phenoxy diphenyl amine, 3′ methyl 2-4-dibutoxy diphenyl amine, p′ ethoxy p′ ethyl diphenyl amine, p-methoxy p′ methyl diphenyl amine, 2-2′ diamino 4-4′ di bromphenyl ether, 2-4-ditoloxy diphenyl amine, 2-4-dihydroxy 4′ methoxy diphenyl amine, N-xylyl phenetidine, 2-4 dihydroxy 4′ ethoxy diphenyl amine, 2-4 dihydroxy 6-methyl 4′ toloxy diphenyl amine, N-naphthyl phenetidine (alpha and beta), 4-4′ ditoloxy 2-2′ dimethyl diphenyl amine, 4-4′ dimethoxy 2-2′ tetra methyldiamino diphenyl amine, 4-4′ dihydroxy 2-2′ diethoxy diphenyl amine, 2-2′ dihydroxy 4-4′ diethoxy diphenyl amine, 4-4′ dihydroxy 2-2′ dimethoxy diphenyl amine, 2-4-diamino 4′ hydroxy phenyl tolyl ether, 4-4′ diphenoxy 2-2′ dichlor diphenyl amine, 2-2′ diamino 4-4′ dichlor diphenyl ether and 2-6-dimethyl 2′6′ diamino diphenyl ether.

Still others are 4-4′ dimethoxy 3-5-dimethoxy diphenyl ether, 4-hydroxy 3-5-dimethoxy dixylyl amine, 4′ ethoxy 3-5-dimethyl diphenyl amine, N N′ dichlorethyl diamino diphenyl ether, 4-hydroxy 4′ amino diphenyl ether, 4-hydroxy 4′ dimethyl amino diphenyl ether, 4-amino 4′ chlor diphenyl ether, 4-4′ diethoxy ditolyl amine, 4-xyloxy phenyl beta naphthyl amine 4-ethoxy phenyl alpha naphthylamine, p-methoxy phenyl 1-8-dimethyl beta naphthylamine, p-hydroxy phenyl 2-7-ethoxy alpha naphthylamine, 4-methoxy 2-methyl phenyl beta naphthylamine, 4-phenoxy 2-6-dimethyl phenyl beta naphthyl amine, 4-phenoxy 2-methyl diphenyl amine, 2-4-6-trimethoxy phenyl alpha naphthyl amine, 2 methoxy (para hydroxy anilino) 1-4-naphthoquinone, 2-methoxy (para toluidino) 1-4-naphthoquinone, 4-amine 4′ tolyl diphenyl ether, and 4-toloxy 4′ butyl diphenylamine.

Also falling within the scope of the invention are 2-2′ amino 4-4′ diethyl diphenyl ether, naphthoxy methyl dimethyl amine (alpha and beta), 4-hydroxy phenyl piperidyl ether, naphthoxy ethyl dimethyl amine, methyl amino diphenyl ether, 2-amino 2′ 4′ dichlor diphenyl ether, phenoxy naphthyl methyl amine, phenoxy naphthyl anthracyl amine, naphthoxy naphthyl dimethyl ether (alpha and beta), 1-8 dimethoxy dibeta naphthyl ether, p-hydroxy p′ methyl amino diphenyl ether, p-hydroxy p′ dimethyl amino diphenyl ether, p-amino phenyl tolyl ether, p-butoxy p′ methyl ditolyl amine, p-butoxy p′ ethyl diphenyl amine, p-methoxy p′ di isopropyl diphenyl amine, 2-nitro 4-amino diphenyl ether, p-methoxy p′ methyl dinaphthylamine, p p′ diamino phenyl xylyl ether, p-hydroxy diphenyl amine, p p′ dihydroxy diphenyl amine.

Methoxy phenyl naphthylol amine (alpha and beta), ethoxy naphthyl phenylol amine (alpha and beta), methoxy naphthyl phenylol amine, (alpha and beta), 7 (tolyl amino) 2-methoxy naphthalene, 7 (xylyl amino) 2-methoxy naphthalene, 2 (tolyl amino) 1-methoxy naphthalene, 2 (xylyl amino) 1-methoxy naphthalene, methoxy phenyl naphthylol nitrosamine (alpha and beta), methoxy phenyl naphthylol methyl amine (alpha and beta), phenylol naphthylol methoxy phenyl amine (alpha and beta), p-butoxy phenyl tolyl amine, p-butoxy phenyl xylylamine, p p′ butoxy diphenyl amine, p-methoxy phenyl tolyl nitrosamine, p-ethoxy phenyl xylyl amine, p-ethoxy phenyl p p′ dinaphthyl amine (alpha and beta) p p′ dimethoxy ditolyl amine, p-methylamino phenyl p′ hydroxy biphenyl ether, p-methylamino phenyl p′ ethoxy biphenyl ether, p-methylamino hydroxy phenyl tolyl ether, chlor phenyl amino diethyl ether, p-phenyl amino chlor phenyl biphenyl ether, p-phenyl amino bromphenyl biphenyl ether.

Also, N-butyl p-amino phenol, N-methyl p-amino phenol, N-isopropyl o-amino m-cresol, N-ethyl o-amino m-cresol, N-diethyl p-amino phenol, N-isobutyl p-amino phenol, N-amyl p-amino catechol, N-isoamyl p-amino phenol, N-cyclohexyl p-amino phenol, p-hydroxy phenyl ethyl amyl amine, N-methyl cyclohexyl p-amino phenol, N-hydroxy butyl m-amino phenol, N-hydroxyethyl m-amino phenol, N-hydroxy propyl m-amino phenol, N-allyl m-amino phenol, p-ethoxy diphenylamine, p p′ diethoxy di tolylamine, o-ethoxy diphenylamine, m-ethoxy diphenylamine, p-ethoxy phenyl naphthylamine, p-ethoxy phenyl tolyl amine, tolyl aminoethyl naphthol, xylyl amino ethyl naphthol, ditolyl dianisidine, dixylyl dianisidine, etc.

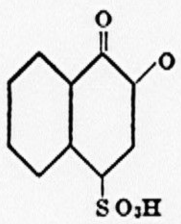
Also, p-ethoxy p′ isopropyl diphenylamine, p-isopropyl phenyl amino di (biphenyl) ether, ethoxy phenyl toluidine, mono naphthyl diamino ditolyl ether, p p′ diethoxy diphenyl nitrosamine, methoxy diphenylamine, amyloxy diphenylamine, butoxy ditolyl amine, phenoxy ditolyl amine, p-tolyl amino p′ dimethyl amino diphenyl ether, dianisyl diamino dimethyl ether, diphenyl diamino diethyl ether, p-tolyl amino diphenyl ether, p-naphthylamino ditolyl ether, p p′ di (tolyl amino) diphenyl ether, o o′ di (tolyl amino) diphenyl ether, diphenylol diamino dibenzyl ether, ditolyl diamino dibutyl ether, dibeta napthyl diamino ditolyl ether, etc.

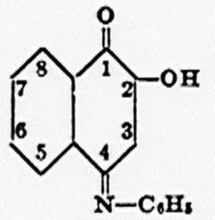
Typical of compounds in which a sulphur, selenium or tellurium atom replaces the oxygen in the compounds mentioned above are p-methyl amino phenyl tolyl selenide, p-methylamino phenyl hydro selenide, p-dimethyl amino tolyl beta naphthyl selenide, p-isopropyl amino phenyl hydro selenide, p-cyclohexyl amino phenyl hydro selenide, p-amino chlorphenyl beta naphthyl selenide, p-methyl amino phenyl tolyl telluride, p-methyl amino phenyl hydro telluride, p-dimethyl amino tolyl beta naphthyl telluride, p-iso amyl amino phenyl hydro telluride, p-cyclohexyl amino phenyl hydro telluride, p-amino chlorphenyl beta naphthyl telluride, etc. It is obvious that in addition to the specific compounds listed above there are selenium and tellurium counterparts for each of the oxygen compounds herein disclosed; also, that there are corresponding sulphur compounds for the various oxygen, selenium and tellurium compounds enumerated.

In general, methods for preparing the antioxidants described above will suggest themselves. For instance, para hydroxy diphenyl amine may be prepared by adding 1 mol. of undissolved hydroquinone to a mixture of 4 mols of aniline and 2 mols of calcium chloride in the absence of any solvents other than the aniline itself. The material is heated for a period of 8 to 10 hours at a temperature of 150–160 degrees C. after which the product, containing para hydroxy diphenyl amine and diphenyl p-phenylene diamine, is steam distilled in order to remove excess aniline. It is then further purified by recrystallizing from benzene or alcohol. The para hydroxy diphenyl amine portion, melting at 69–70 degrees C., is obtained in good yield.

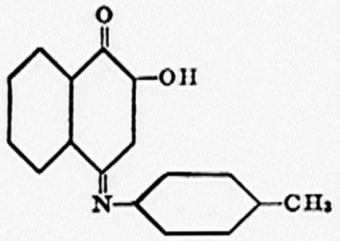
Para para' dihydroxy diphenyl amine may be prepared similarly by reacting in the absence of solvents equimolecular quantities of hydroquinone, para amino phenol and calcium chloride in an autoclave for a period varying from 5 to 8 hours at a temperature of approximately 175 degrees C. The product, para para' dihydroxy diphenyl amine, is treated with a little hydrochloric acid and boiled with water after which it is passed through a filter in order to remove any undissolved residues and is then precipitated by the addition of an excess of common salt. The precipitate then dried is in the form of a bluish powder melting at a temperature ranging from 170–180 degrees C.

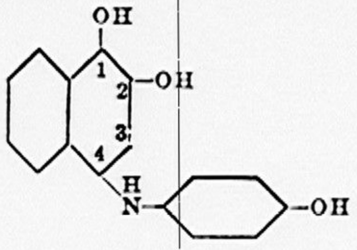
Another method of preparation of certain of these materials is that outlined in Berichte (1924), volume 27, page 23. In accordance with this method 1-amino 2-naphthol 4-sulfonic acid is obtained by treating alpho nitroso beta naphthol with a saturated solution of sodium bisulfite. This reaction causes a simultaneous reduction of the nitroso group to an amino group and the introduction of a sulfonic group in the 4-position of the naphthyl radical. The sulfonate thus formed may readily be oxidized by means of nitric acid to form 1-2-naphthoquinone 4-sulfonic acid which upon being treated with potassium chloride forms a potassium salt. The potassium salt is then reacted in mol. per mol. ratio with aniline in aqueous solution, whereupon an anilido group is substituted for the sulfonate radical and 2-hydroxy 1-4-anilido naphthoquinone, results. This compound is formed as a red precipitate, which upon being filtered from the reacting materials, gives a solid material having a melting point of about 240 degrees C., a compound in relatively pure form. For most purposes it is unnecessary to subject it to further purification.





Similarly, if in the above reaction para toluidine is substituted for aniline, the compound 2-hydroxy para toluidino 1-4-naphthoquinone having the formula is obtained. Likewise if para amino phenol is substituted for aniline, 2-hydroxy para hydroxy anilino 1-4-naphthoquinone is formed. Also, the compound 1-2-dihydroxy 4-para hydroxy phenyl naphthylamine is obtained by interacting the potassium salt of 1-2-naphthoquinone 4-sulfonic acid with para amino phenol in the presence of an active reducing agent such as sodium hydrosulfite.





Others of the foregoing compounds may, of course, be prepared according to still other methods. Thus n-butyl p-amino phenol can be prepared by reacting butyl amine and p-chloro phenol in an autoclave; p-ethoxy phenyl alpha naphthyl amine, by reacting p-phenetidine with alpha naphthol; n-isopropyl o-amino m-cresol, by interacting o-amino m-cresol, acetone and formic acid; and naphthoxy methyl dimethyl amine, by treating beta naphthol dissolved in alcohol with dimethyl amine and formalin. Still other methods which it is not necessary to describe in this application may be employed where circumstances permit, such methods being found in the literature of chemistry.

It is to be understood that the invention is not limited to any specific method of preparing the antioxidants or, regardless of illustrative references herein, to rubber. The antioxidants of the invention may be employed advantageously in substantially any of the standard rubber formulae, the following being one in which they have been found to give excellent results.

	Parts
Extracted pale crepe	100
Zinc oxide	5
Hexamethylene tetramine	1
Sulfur	3
Stearic acid	1½
Antioxidant	1

As pointed out above, they may also be employed in transformer oils, gasoline, etc.

Rubber samples prepared in accordance with the foregoing formula were subjected to vulcanization for varying periods of time and then tested to ascertain their physical characteristics. One set from each type of stock was tested immediately for tensile strength and elasticity while a second set was subjected to ageing in a Bierer-Davis bomb for 6 days in oxygen at 50 degrees C. and 150 pounds per square inch. At the conclusion of the 6 day period the samples were removed from the bomb and subjected to physical tests corresponding to those conducted upon the unaged samples.

It is evident from the following tables that rubber compositions containing even small proportions of the antioxidants of this invention resist deterioration remarkably well. Similar compositions not containing the antioxidants upon being subjected to corresponding tests increase in weight in the neighborhood of 10% and lose almost all their tensile and elongation properties. In the following data the tests for para para' dihydroxy diphenyl amine and para hydroxy diphenyl amine were obtained from a rubber formula similar to that given above with the exception that the sulfur content was increased from 3 to 4 parts.

Cure in mins. 40#	Original				Aged				Pct. wt. inc.
	Ult. tens.	Max. elg.	Load in kgs/cm²		Ult. tens.	Max. elg.	Load kgs/cm²		
			500%	700%			500%	700%	
Para hydroxy diphenylamine									
30	107	870	44	------	132	810	72	------	0.0
50	170	840	78	------	182	770	126	------	0.0
70	186	790	110	------	178	725	155	------	0.17
Para para' dihydroxy diphenylamine									
50	125	835	20	62	140	815	23	74	0.3
70	160	815	26	86	145	765	31	103	0.4
2-hydroxy para toluidino 1-4-naphthoquinone									
35	91	870	13	36	91	820	15	46	0.14
50	101	800	17	55	118	795	20	64	0.28
70	139	775	23	86	128	725	28	104	0.30
2-hydroxy (para hydroxy anilino) 1-4-naphthoquinone									
35	82	875	12	33	108	840	16	48	0.01
50	109	840	16	49	121	810	19	64	0.03
70	133	780	24	53	129	745	26	96	0.05
1-2-dihydroxy 4-(para hydroxy phenyl) naphthylamine									
35	80	900	10	27	102	865	15	42	0.02
50	110	840	14	48	122	805	20	66	0.09
70	134	795	21	76	138	760	25	95	0.11
2-methyl 4-hydroxy diphenylamine									
35	115	880	14	42	120	845	16	53	0.02
50	136	820	20	66	132	785	21	79	0.09
70	159	795	24	87	151	750	28	112	0.06
2-hydroxy 1-4-anilido naphthoquinone									
30	100	900	11	32	85	815	14	44	0.20
50	135	865	17	54	125	790	21	75	0.28
70	165	805	24	90	130	730	29	111	0.54

Similar data have been obtained in the case of others of the compounds mentioned, for example, naphthoxy methyl dimethyl amine (dimethyl amino methyl beta naphthyl ether) and naphthoxy methyl diethyl amine (diethyl amino methyl beta naphthyl ether). Using the same formula, but with 4 parts of sulfur rather than 3 parts, the following results were obtained:

Cure	Original				Aged				Percent wt. inc.
	Tens.	Elg.	500%	700%	Tens.	Elg.	500%	700%	
Dimethyl amino methyl beta naphthyl ether									
30/285	80	830	13	39	45	730	13	37	.87
50	150	815	22	77	60	655	25	-----	1.17
110	165	750	31	120	90	650	37	-----	1.55
Diethyl amino methyl beta naphthyl ether									
30/285	110	850	16	48	65	740	17	51	.55
50	175	805	27	96	100	705	29	97	.81
110	185	735	39	150	120	675	42	-----	1.37

The following data came from similar tests on N-butyl p-amino phenol, N-isopropyl, o-amino m-cresol, N-methyl p-amino phenol, N-tolyl phenetidine and N-phenyl phenetidine, using the formula as first given:

Cure	Original				Aged				Percent wt. inc.
	Tens.	Elg.	500%	700%	Tens.	Elg.	500%	700%	
N-butylamino phenol									
35/285	162	840	19	20	144	780	23	91	.06
50	165	765	27	112	134	725	29	117	.31
70	186	725	38	160	140	765	41	-----	.53
N-isopropyl o-amino m-cresol									
35/285	115	850	14	47	126	745	26	96	.71
50	140	780	23	85	143	680	40	-----	.19
70	161	755	28	112	152	645	55	-----	.28
N-methyl p-amino phenol									
35/285	157	740	33	129	164	685	47	-----	.27
50	176	705	44	169	170	650	60	-----	.96
70	203	685	56	-----	186	635	74	-----	.30
N-tolyl phenetidine									
35/285	112	915	12	32	89	850	13	36	.04
50	116	825	17	55	122	840	17	55	.17
70	122	780	20	76	123	770	22	79	.22
N-phenyl phenetidine									
35/285	110	875	14	42	108	845	14	45	.20
50	119	805	18	60	105	785	18	62	.23
70	132	780	22	78	129	745	25	97	.27

From the foregoing it is evident that the compounds herein disclosed are highly suitable as age resisters of rubber and other organic products which undergo deterioration under the influences of heat, light and oxygen. Not only do the compounds of the present invention counteract the effects of such influences, but they tend to impart other highly desirable qualities, such, for example, as increased resistance to deterioration by flexing.

It is to be understood that as hereinafter used the term "benzenoid" embraces phenyl and naphthyl groups, which, as preferred, do or do not contain substituents, such substituents being, for example, alkyl, alkoxy, amino and like groups. Also, by the term "rubber" it is meant to include rubber, latex, balata, gutta percha, guayule, rubber isomers, rubber conversion products and similar materials. It will be apparent that numerous changes may be made in the procedure to be followed and the chemicals employed without departing from the inventive concept. It is intended that the patent shall cover, by suitable expression in the appended claims, whatever features of patentable novelty reside in the invention herein disclosed.

What I claim is:

1. The method of preserving rubber which comprises treating the same with p-ethoxy phenyl naphthyl nitrosamine.

2. A rubber composition comprising rubber and p-ethoxy phenyl naphthyl nitrosamine.

ALBERT M. CLIFFORD.

Deft. Ex. No. 2-F

BRITISH
PATENT SPECIFICATION

350,438

Application Date: March 5, 1930. No. 7276/30.

Complete Accepted: June 5, 1931.

COMPLETE SPECIFICATION.

Improvements in or relating to Motor Fuels.

(A communication from **STANDARD OIL COMPANY**, a Corporation organized under the laws of the State of Indiana, of Chicago, Illinois, and of Whiting, Indiana, United States of America.)

I, **HAROLD WADE**, a British Subject, of 111/112, Hatton Garden, London, E.C. 1, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

The present invention relates to improvements in hydrocarbon motor fuel products of the character of gasoline boiling within the range of approximately 80—450° F., and has to do more particularly with motor fuel products derived by the cracking of higher boiling hydrocarbon oils and which display a tendency to form gum or resinification products on storage, even after refining treatment with a dilute sulphuric acid such as sulphuric acid of 75% strength or lower. This tendency may be indicated by an accelerated test designated hereinafter as the oxygen absorption test, which is a measure of the potential gum formation or resinification properties of the motor fuel products.

In methods of cracking high boiling hydrocarbon oils to produce lower boiling hydrocarbon oils, the products which have been secured may in general be grouped in two classes; the first, which at the present time forms the bulk of pressure cracked gasoline, having an anti-knock value somewhat greater but not substantially greater than straight-run gasoline; a somewhat lower A.P.I. gravity than straight-run gasoline, containing a moderate proportion of unsaturated compounds as determined by combined absorption and polymerization by sulphuric acid in accordance with the method of determination hereafter set forth; and being in general of such a character that the constituents thereof which tend to form gum on storage and handling are readily removed by treatment with sulphuric acid to produce a product comparable in that respect to straight-run gasoline. Motor fuel products of the second general class hitherto derived principally from vapour phase processes or very high temperature pressure cracking processes, have anti-knock values substantially higher than straight-run gasoline; have a much lower A.P.I. gravity; contain large amounts of unsaturated hydrocarbons as determined by sulphuric acid absorption and polymerization; and require excessive treatment with sulphuric acid, with very substantial losses, to remove potential gum-forming constituents.

Although it cannot be said that there is a hard and fast line between the two types of products, the former type of product, with 425° F. end point will in general have an anti-knock value not exceeding an equivalent of 1.2 cubic centimeters of tetra-ethyl lead per gallon of straight-run Mid-Continent gasoline; an olefine content as determined by sulphuric acid absorption and polymerization of not over 40%; an A.P.I. gravity of 56 to 59°; and an oxygen absorption induction period greater than 125 minutes, as determined by the test hereafter set forth. The second type of product, which may be designated as the vapour phase type, will in general have an anti-knock value equivalent to at least 1.2 cc. tetra-ethyl lead per gallon straight-run Mid-Continent gasoline, an A.P.I. gravity of about 49 to 55°; an unsaturated content as determined by sulphuric acid absorption and polymerization exceeding 40%, and usually above 45%; and an oxygen absorption induction period of less than 125 minutes.

The latter type of product in general tends to form gum, even on storage in the dark, and under like conditions, tends to lose its anti-knock properties. The same tendencies may exist, to a greater or less extent, in products of the first-mentioned type and the present invention may be employed in connection therewith, although particularly intended for use in connection with products of the so-called vapour-phase cracked type. Thus, products of certain processes commonly designated as liquid phase or liquid-vapour phase, although having a lower unsatur-

[*Price* 1/-]

ated content than vapour phase products, as shown by combined absorption and polymerization by sulphuric acid, have gum-forming properties similar to those of vapour phase products. The present invention may be applied in connection with such process, as well as with such highly cracked motor fuels as are prepared by drastic or high temperature cracking for destructive distillation of bituminous material, such as coal, peat, lignite, and the like.

In determining the oxygen absorption induction period, by which an indication of the potential gum-forming properties of the gasoline on storage and handling may be secured, the following procedure may be employed: 100 cubic centimeters of the motor fuel or gasoline to be tested are placed in a one litre flask, the total volumetric capacity of which is 1150 to 1200 cubic centimeters. The gasoline is maintained at about 212° F. by heating in a steam bath while an oxygen atmosphere is maintained in the flask under a substantially constant pressure of about 2½ atmospheres. The flask is vigorously agitated and the volume of oxygen absorbed is measured at frequent intervals. The oxygen absorption is characterized by an initial period in which relatively small amounts are absorbed and subsequent to this period there is a rapid increase in the rate of absorption. This initial period of slow absorption is designated the induction period and its end is regarded as the point at which oxygen absorption under the conditions stated reaches a rate of 1 cubic centimeter per minute. If the oxygen absorption is plotted against the time in minutes with equal distances on the coordinates for time in minutes and absorption in cubic centimeters, it will be evident that the end of the induction period will be indicated upon the curve by the point where the tangent to the curve has a slope of 45°. The length of the induction period, as determined by this test, provides a fairly accurate indication of the potential gum-forming behaviour of the motor fuel product on storage and handling.

A typical vapour-phase cracked distillate having a 425° F. end point will have a gravity of 48 to 56° Baumé as compared with 58 to 60 Baumé for a straight-run gasoline of similar boiling range from Mid-Continent crude oil. The product, without treating, is found to have an oxygen absorption induction period of about 35 to 70 minutes. It is suitably treated with dilute sulphuric acid, say 60% acid, to the extent of 6 lbs. of such acid per barrel of distillate. The loss on treating is about ½%, the colour of the product is not substantially altered, but is substantially stabilized, and the oxygen absorption induction period is increased to 80 to 90 minutes. In determining the unsaturated hydrocarbons in such a product it shows a combined absorption and polymerization loss of 50% to 70% when the test is made with 90% sulphuric acid. This loss is determined by subjecting a given volume of the material to treatment at room temperature with two volumes of 90% sulphuric acid, separating the remaining oil from the acid layer and redistilling the unabsorbed oil to produce a distillate having the same end point as the original material treated. The loss in volume relative to the original material is that which is designated as the combined absorption and polymerization loss.

A treated distillate product, produced as hereinbefore described, or other cracked products having a tendency to form gum or resinification products on storage, is treated by adding thereto a small proportion of a substituted amino-phenol having reducing properties. Such substances are employed in proportions of, in general, less than 0.1% and preferably less than 0.05%, and substantially stabilize the product against gum formation or resinification, and also against loss in anti-knock. This stabilization is indicated by an increase in the oxygen absorption induction period of the products to which they are added. As examples of such materials, we have employed para- and ortho-methyl-amino-phenol, dimethyl-amino-phenol, butyl, isopropyl and benzyl amino-phenols, dihydroxy-diphenyl-amine, and the like.

The specific gravity of these materials varies to a considerable extent, all of them, however, being effective in inhibiting the continued development of gum in the motor fuel product and preventing its deterioration, for example, by decrease of its anti-knock value, and these properties are indicated by an increase in the induction period of the product. It is preferred to employ those which, when added to a vapour-phase type of product as hereinbefore set forth, in proportion of 0.05% or less, will increase the induction period to above 125 minutes and preferably to 300 minutes or more. Thus, with the product prepared as above described, and which, after dilute acid treatment has an induction period of 55 minutes and a lead equivalent of 1.7% cubic centimeters tetra-ethyl lead per gallon of straight-run Mid-Continent gasoline, the addition of 0.01% of para-methyl-amino-phenol increases the induction period to 1800 minutes and 0.001% to 435 minutes.

The results secured by the application

of the oxygen absorption induction test, as hereinbefore described, definitely indicate the behaviour of the motor fuel product, both in its tendency to form gum constituents and to lose in anti-knock value. Thus, two lots of the same gasoline were stored for eight months, one without added constituents and the other with 0.002% of para-methylaminophenol added. At the end of eight months, the untreated gasoline showed a gum test of 42 milligrams of non-volatile resin per 100 cc. whereas the lot treated with para-methylaminophenol showed the formation of only 2 milligrams. Similarly, samples of gasoline produced by cracking Mid-Continent gas oil in vapour phase and having an anti-knock value equivalent to 1.7 cubic centimeters of tetra-ethyl lead per gallon of Mid-Continent gasoline, without adding constituents, at the end of seven months, had an anti-knock equivalent of zero; that is, no higher than Mid-Continent gasoline. The same product, to which 0.002% of para-methylaminophenol was added, and which was stored under similar conditions for the same period of time, showed at the end of that period an anti-knock value of 1.5 cubic centimeters or only slightly under the original anti-knock value had when placed in storage.

The marked extension of the oxygen absorption induction period by the addition of the substances having the characteristics of the bodies hereinbefore described is not accompanied by any change in the anti-knock value, 90% acid absorption and polymerization loss or other characteristic property of the motor fuel.

Motor fuels produced in accordance with the present invention may be recognized by the fact that their oxygen absorption induction period is decreased by successive treatments with dilute acid and dilute alkali. In the preferred form of the invention, in which the motor fuel product shows a loss of 40% or higher by absorption and polymerization by 90% sulphuric acid, this decrease may be from upwards of 125 minutes, and generally upwards of 300 minutes to less than 100 minutes. Thus, a vapour-phase pressure distillate product having an anti-knock value equivalent to about 2 cubic centimeters of tetra-ethyl lead per gallon of straight-run gasoline and showing a loss on absorption and polymerization by 90% sulphuric acid exceeding 50%, had an induction period of approximately 45. After treatment with 6 lbs. per barrel of 60% sulphuric acid, the anti-knock value and sulphuric acid absorption and polymerization loss were substantially unchanged and the induction period increased to approximately 86 minutes. The addition of 0.002% of para-methylamino-phenol increased the induction period to about 520 minutes without affecting the other characteristics of the motor fuel. This product constitutes a preferred embodiment of the present invention. On washing a given volume of it successively with 10% by volume of 50% sulphuric acid or other dilute sulphuric acid of insufficient strength to effect treating, and 10% by volume, of approximately 10% alcoholic sodium hydroxide solution, the oxygen absorption characteristics were altered, the induction period being reduced to about 58 minutes.

If desired, the anti-knock value of the motor fuel described herein may be increased by the addition of small amounts of tetra-ethyl lead or other suitable material and it is intended that the accompanying claims shall include the products of the type herein set forth regardless of the addition thereto of tetra-ethyl lead or other materials modifying their fuel characteristics.

We are aware that it has already been proposed to add small quantities of amino-phenol to motor fuels for the purpose of inhibiting resinification. The substituted amino-phenols which are used in the present invention are very much more effective than the amino-phenols, the quantitative efficiency of for example paramethyl amino-phenol being of the order of eight times that of para- or ortho-aminophenol.

Having now particularly described and ascertained the nature of my said invention, and in what manner the same is to be performed, as communicated to me by my foreign correspondents, I declare that what I claim is:—

1. As a composition of matter, a cracked hydrocarbon motor fuel distillate normally tending to deteriorate and develop gum on storage, said distillate containing a small proportion of an added compound comprising a substituted amino-phenol.

2. A composition of matter according to claim 1, wherein substituted amino-phenol is in a proportion of less than 0.05%.

3. A composition of matter according to claim 1, wherein the distillate is a vapour-phase cracked product.

4. A composition of matter according to claim 1 or 3, wherein the added compound is a methylaminophenol.

5. A composition of matter according to claim 1 or 3 wherein the added compound is a benzyl-amino-phenol.

6. The method of preventing deterioration of and inhibiting the development of gum-forming constituents in cracked motor fuel distillates and particularly vapour-phase cracked distillates which consists in adding thereto a small proportion of a compound comprising a substituted amino-phenol.

7. The method in accordance with claim 6, wherein the substituted amino-phenol is added to the amount of not more than 0.05%.

8. The method in accordance with claim 6, wherein the added compound is methyl-amino-phenol.

9. The method in accordance with claim 6, wherein the added compound is benzyl-amino-phenol.

10. The motor fuel and the method of preparing the same substantially as described.

Dated this 4th day of March, 1930.

BOULT, WADE & TENNANT,
111/112, Hatton Garden, London,
E.C. 1,
Chartered Patent Agents.

Redhill: Printed for His Majesty's Stationery Office, by Love & Malcomson, Ltd.—1931

Deft. Ex. No. 2-G

Division 6
Room 4898-C
Lidoff:GV

Paper #14

DEPARTMENT OF COMMERCE

U. S. Patent Office

Washington.

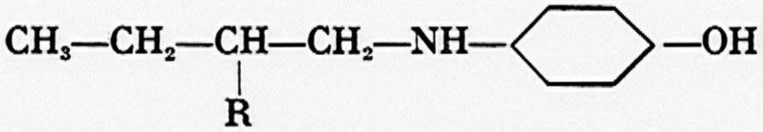
In re Application of
Frederic R. Bean
Serial No. 134,903
Filed April 3, 1937
For: N-substituted Aminophenols and Motor Fuels Stabilized therewith.

On Appeal
Before the Board of Appeals.

Examiner's Statement.

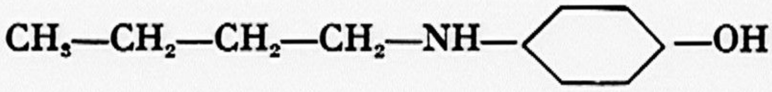
This is an appeal from the final rejection of claims 28 and 29. The remaining claims in the application, claims 9 to 14, 26 and 27 are allowable as far as is now known. The finally rejected claims read as follow:

Claim 28. A compound having the formula:



wherein R is a member from the group consisting of hydrogen and an alkyl group of not more than two carbon atoms.

Claim 29. A compound having the formula:



The References are

Downing et al	2,009,767	July 30, 1935
Dahlen et al	2,063,151 (Filed Feb. 6, 1934)	Dec. 8, 1936
Atwell	2,081,130 (Filed Dec. 30, 1932)	May 25, 1937
Rogers et al	2,084,977	June 22, 1937
Clifford	2,109,164 (Filed Oct. 22, 1932)	Feb. 22, 1938
British patent	350,438	June 5, 1931

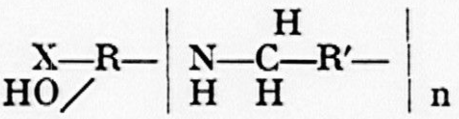
The Invention.

The invention relates to the formation of N-alkyl P-aminophenols by reduction of the reaction product of the phenol and the corresponding aldehyde. The claims drawn to this process (9 to 14, 26 and 27) are allowable. The claims are also drawn to a specific product, N-n-butyl p-aminophenol (claim 29) and to a generic product claim (claim 28) drawn to certain N-alkyl P-aminophenols.

The References.

Downing et al disclose a process for reducing aralkylidene and *alkylidene* aminophenols to the corresponding N-aralkyl and N-alkyl aminophenols. One of the starting materials disclosed by Downing et al is "butyral-4-aminophenol, the reduction of which, in the Downing et al process, will produce butyl-4-aminophenol.

Dahlen et al also disclose the formation of N-alkyl aminophenols by reducing the corresponding N-alkylidene compound. N-heptyl-para aminophenol is specifically disclosed and a generic formula is disclosed of compounds of the type



$$\begin{array}{l}X-R-\\HO/\end{array}\left|\begin{array}{l}\quad\;\;H\\N-C-R'-\\H\quad H\end{array}\right|_n$$

wherein X may be hydrogen, R is benzene, R′ is *alkyl* and n is an integer.

Rogers et al, which was inadvertently dropped as being overcome by applicant's Rule 75 Affidavit, is relied upon since it is entitled to a date of February 10, 1930 as to the disclosure of "para — butyl aminophenol" (see page 8 of the parent case SN 427,416).

Atwell discloses "butylaminophenol", and "4-n-amylaminophenol".

Clifford discloses "N-butyl P-aminophenol" "N-isobutyl P-aminophenol", "N-isoamyl P-aminophenol".

The British patent disclosed "para — butyl aminophenol".

The Rejection

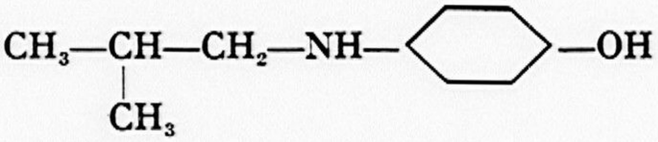
The allowed claims, being drawn to process, are patentably distinct from the rejected product claims.

Claims 28 and 29 are rejected as being unpatentable over each of the cited patents. In a companion case serial number 175,078 of the same applicant, claims to a motor fuel comprising hydrocarbons and the above claimed compounds were held, in a decision upon appeal docket No. *32,681*, to be directly anticipated by each of the above cited British patent, Rogers et al and Clifford, upon the ground that the term "butyl" unless otherwise identified by a prefix means *normal* primary butyl and that hence the compounds disclosed by these patents are identically the same as the claimed compound. This is clearly illustrated by Clifford, who discloses "N-butyl-P-aminophenol" and then when he wishes to designate a *different* butyl group, specifies "N-*iso*butyl P-aminophenol. The same argument is applicable to Downing et al since the term "butyral" means the *normal* radical unless otherwise specified, and hence the compound obtained by Downing et al in his reduction process must be N- normal primary butyl P-aminophenol, the claimed product. The same argument also applies to the Atwell disclosure of "butylaminophenol". In view of the decision of the Board of Appeals in the companion case, these references fully meet the product claims.

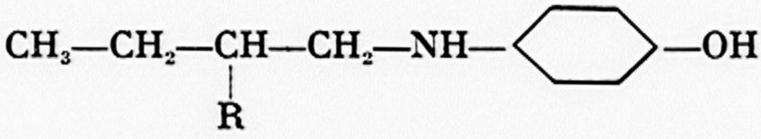
The product claims are further rejected as lacking in invention over Dahlen et al who disclose a higher homologue of the claimed compounds and who disclose a generic formula which includes the claimed compound. There is no invention seen in picking out one of the products included in the general formula of Dahlen et al (See The Hamilton Laboratories Inc. v. Massengill 45 U. S. P. Q. 594).

The claims are further rejected as lacking in invention over Atwell, who clearly discloses "4-n-amylaminophenol" the next higher homologue of the claimed product having one more carbon atom in the alkyl chain. The disclosed compound obviously contains a straight chain alkyl group of 5 carbon atoms attached to the amino group para to the hydroxy group on the benzene ring. There is no showing that the claimed product possesses any properties not exhibited by the higher homologues of the prior art.

The product claims are further rejected as lacking in invention over the Clifford disclosure of "N-isoamyl P-aminophenol" a compound which is a higher homologue and an isomer of the claimed compounds (claim 28) and a higher homologue of the product of claim 29. This compound, disclosed by Clifford, differs from those claimed in having a methyl group in the 3 position of an n-butyl chain, that is to say the Clifford compound is



whereas applicant's products are



where R is —H, —CH_3 or —C_2H_5. It can be seen that Clifford's compound is a next higher homologue of the n-butyl derivative and is merely a position isomer of the substituted products of claim 28. The compound contains the elements, essential to the claimed products, that is a straight chain group of four carbon atoms attached to the nitrogen atom of a *para* amino phenol. The compound differs only in having a non-reactive substituent in the 3 position in the chain instead of having no substituents or the *same* substituent in the 2 position in the chain.

The compounds of Clifford, Rogers et al, and Dahlen et al are disclosed to be useful as gasoline antioxidants, the same purpose as that for which applicant's compounds are employed.

It is submitted that the final rejection is sound and should be affirmed.

Respectfully,

Examiner,
Division 6.

Newton M. Perrins
c/o Eastman Kodak Company
343 State Street
Rochester, New York.

Deft. Ex. No. 2-H

Appeal No. 37,498 **MM**
Hearing:
April 22, 1941

IN THE UNITED STATES PATENT OFFICE

BEFORE THE BOARD OF APPEALS

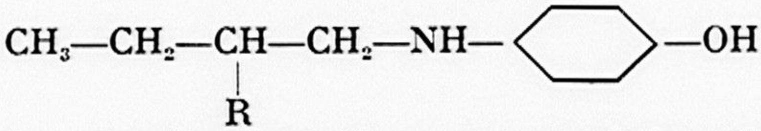
Ex parte Frederic R. Bean

Application for patent filed April 3, 1937, Serial No. 134,903. N-Substituted Aminophenols and Motor Fuels Stabilized Therewith.

Messrs. Newton M. Perrins & Daniel I. Mayne for applicant.

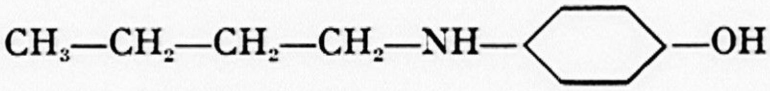
This is an appeal from the decision of the examiner finally rejecting the following claims:

28. A compound having the formula:



wherein R is a member from the group consisting of hydrogen and an alkyl group of not more than two carbon atoms.

29. A compound having the formula:



The references relied on are:

British patent	350,438	June 5, 1931
Downing et al	2,009,767	July 30, 1935
Dahlen et al	2,063,151	Dec. 8, 1936
Atwell	2,081,130	May 25, 1937
Rogers et al	2,084,977	June 22, 1937
Clifford	2,109,164	Feb. 22, 1938

The appealed claims cover a composition which is said to be of value as an antioxidant for gasoline. Claim 28 is for N-(normal primary butyl)-p-aminophenol, while claim 29 is broader in that it covers a restricted class of N-alkyl-p-aminophenols.

The examiner has called our attention to the fact that appellant had another application, Serial No. 175,078, filed November 17, 1937 on which an appeal was taken and decided by this Board March 29, 1940. Claim 2 of said other application reads as follows:

> 2. A motor fuel comprising hydrocarbons and normally tending to deteriorate containing, in an amount sufficient to substantially inhibit the deterioration, N-(n-primary-butyl)-p-aminophenol.

This claim differs from those before us at the present time in that it calls for a motor fuel containing the compound claimed here. This claim stood rejected on the British patent

No. 350,438 which is also a reference in the present case. In discussing this claim 2, this Board said—

> "Considering the terms of claim 2, the preservative is required to be "N-(n-primarybutyl)-p-aminophenol". It is clear that this identifies the definite chemical compound where the butyl radical must be normal primarybutyl and must be attached specifically to the nitrogen of the amine residue and the substituted amine must stand in the para position with respect to the hydroxyl.
>
> It is the examiner's view that the British patent to Wade No. 350,438 in specifying employment of parabutyl-aminophenol in this relation is a direct anticipation of claim 2 and others in this respect. We are in full agreement with the examiner's holding that unless otherwise specified the above term of the British patent means identically the same compound only more fully described by applicant's term in claim 2. We see no reason for doubt that as to the first point, if not otherwise described, the simple term "butyl" indicates as universally understood, the simplest form of the butyl radical normal primarybutyl. If iso, secondary or tertiary forms are intended, these are prefixed to the butyl. We agree further with the examiner's conclusion that unless otherwise specified the term of the British patent also means that the butyl radical is attached to the nitrogen of the amine residue otherwise the term "para" would have no reasonable meaning. If the butyl and amine residues should stand at different positions on the benzene ring with respect to the hydroxyl both positions would have to be specified. It would be unreasonable to assume that the para position might be intended to apply uncertainly to one or the other of the positions of the two substituents."

Since the claim before us differs so little from those previously considered by us, our reasons for affirming the examiner apply as well to this case as it did to the other. We can see no good reason for changing our opinion.

It is not deemed necessary to discuss the other references in detail since they are deemed to be accumulative and not so pertinent as the British patent noted above.

The decision of the examiner is affirmed.

W. L. Redrow Examiner-in-Chief F. P. Edinburg Examiner-in-Chief C. H. Shaffer Examiner-in-Chief	BOARD OF APPEALS

Mr. Newton M. Perrins,
c/o Eastman Kodak Co.,
343 State St.,
Rochester, N. Y.

May 7, 1941

Deft. Ex. No. 2-I

Appeal No. 37,498 MM

IN THE UNITED STATES PATENT OFFICE

BEFORE THE BOARD OF APPEALS

Ex parte Frederic R. Bean

Application for patent filed April 3, 1937, Serial No. 134,903. N-Substituted Aminophenols and Motor Fuels Stabilized Therewith.

Messrs. Newton M. Perrins & Daniel I. Mayne for applicant.

ON PETITION FOR RECONSIDERATION

This is a petition for reconsideration of our decision of May 7, 1941.

The two claims in issue define a chemical compound known as amino phenols. It is alleged that they are useful as antioxidants for gasoline. The examiner rejected the claims as reading directly on British patent No. 350,438 listed above, and this action was affirmed by us.

Apparently appellant in this petition concedes that this compound is mentioned in this patent as an antioxidant for gasoline but contends there is no evidence in the patent that the compounds have ever been made and hence this patent is not an anticipation.

This line of reasoning does not appear to be sound. The patent states "we have employed para —xx butyl x x aminophenols" etc. which would indicate that the compound was actually tried out. In addition, this record shows that substituted amino-phenols as a class are known to have antioxidation properties.

The court decision cited in the brief relates to the composition of cyanine dyes used extensively with photographic emulsions. The dyes are comparatively new and are quite complex. It is not seen that such decision is controlling in this case.

The petition has been granted to the extent indicated but we again affirm the decision of the examiner.

W. L. Redrow
Examiner-in-Chief
F. P. Edinburg
Examiner-in-Chief
C. H. Shaffer
Examiner-in-Chief

BOARD
OF
APPEALS

Mr. Newton M. Perrins,
c/o Eastman Kodak Co.,
343 State St.,
Rochester, N. Y.

July 1, 1941

Patented June 27, 1939 Deft. Ex. No. 3 **2,163,639**

UNITED STATES PATENT OFFICE

2,163,639

COMPOSITION OF MATTER PRODUCED FROM POLYHYDRIC PHENOLS AND ALKYL AMINES

Harold Von Bramer and Albert C. Ruggles, Kingsport, Tenn., assignors, by mesne assignments, to Eastman Kodak Company, Jersey City, N. J., a corporation of New Jersey

No Drawing. Application August 17, 1936, Serial No. 96,466

19 Claims. (Cl. 260—128)

This invention relates to new compositions of matter and more particularly to compositions of matter comprising reaction products of polyhydric phenols and primary or secondary alkyl primary amines and a process for the preparation thereof.

The reaction of methyl amine and hydroquinone has been described in German Patent 260,234 and in U. S. Patent 1,297,685. These patents are concerned with the production of N-methyl-p-aminophenol widely used, in the form of its sulphate, as a photographic developer, and, variously known in the trades as "Metol", "Elon", etc. Our copending application, Serial No. 697,527, filed Nov. 10, 1933, describes a method of preparing substantially pure N-methyl-p-aminophenol from methyl amine and hydroquinone.

Now, we have found that a polyhydric phenol containing at least two phenolic hydroxyl groups in the ortho or para position to one another can be reacted with primary or secondary alkyl primary amines containing at least four carbon atoms, with the elimination of water, to give complex compositions which analyses indicate are made up principally of N-alkylaminophenols together with attendant reaction products of higher molecular weight than the aminophenols. Ordinarily one of these attendant reaction products is apparently a N,N′-dialkylphenylenediamine. Whether our new reaction products are strictly mixtures or whether the ingredients are combined in chemical combination is not known. However, the reaction products do not behave as mixtures in that it has proven impossible to separate the various ingredients by fractional distillation, or, to separate the aminophenols from the reaction products by ordinary chemical means.

We have found that these new complex reaction products are excellent motor fuel stabilizers, surpassing the best known motor fuel stabilizers in several respects. Our new reaction products are characterized by their greater solubility in motor fuels consisting mainly of hydrocarbons, by their greater resistance to extraction by water and/or alkalies therefrom and by their decreased tendency to impart or induce color and/or turbidity therein, as compared with such well known motor fuel stabilizers as N-methyl-p-aminophenol and N-benzyl-p-aminophenol. These new reaction products are from about 4 to 7 times as soluble in motor fuels as N-benzyl-p-aminophenol and from about 120 to 190 times as soluble in motor fuels as N-methyl-p-aminophenol, from about 20 to 30 times as resistant to extraction by water from motor fuels as N-benzyl-p-aminophenol and from about 50 to about 75 times as resistant as N-methyl-p-aminophenol. These new reaction products are nearly twice as resistant to extraction by alkalies from motor fuel as either N-benzyl- or N-methyl-p-aminophenol. These new reaction products are more soluble in methanol than either N-benzyl or N-methyl-p-aminophenol. Motor fuels stabilized with these new reaction products, when allowed to stand in contact with water and iron undergo no discoloration and acquire no turbidity in six days as contrasted with N-methyl-p-aminophenol and N-benzyl-p-aminophenol each of which induces a brown color in the water layer after a few hours. Motor fuels stabilized with these new reaction products show no color or haze upon being exposed to direct sunlight for three hours as contrasted with motor fuels stabilized with N-methyl-p-aminophenol which show a slight haze and a yellow color after about one hour exposure and motor fuels stabilized with N-benzyl-p-aminophenol which show a yellow color and slight haze after about three hours exposure. Motor fuels stabilized with these new reaction products are described in our copending application, Serial No. 96,467, filed August 17, 1936.

In preparing our new reaction products, the primary or secondary amine is reacted with the polyhydric phenol under such conditions that water is eliminated between the phenol and the amine. We have found such conditions are advantageously an excess of amine and the substantial absence of diluents in the reaction mixture other than those formed during the reaction. Ordinarily from about one and one-quarter to about two and one-half molecular proportions of amine to each mole of polyhydric phenol are suitable. Proportions outside these advantageous limits can be used, however, as the molecular proportions of amine are increased above two and one-half the reaction products will be found to become less and less potent as motor fuel stabilizers, i. e., their gum inhibiting properties will be found to become less and less. As the molecular proportions of amine are decreased from one and one-quarter, the reaction products will be found to become less and less potent as motor fuel stabilizers, less and less soluble in the motor fuels, less and less resistant to extraction by water and/or alkalies therefrom, etc. Ordinarily no diluent other than the impurities found in the usual commercial amine (about 10% by weight or less of inert materials, such as water, alcohols, etc.) should be used. As the reaction mixture is diluted from this advantageous concentration, the reaction products will be found to become less and less soluble in motor fuels, less and less resistant to extraction therefrom, etc.

To effect a reaction, the polyhydric phenol and amine, the phenol advantageously being in solution in the excess amine, is heated, advantageously in a closed system such as an autoclave, at about 200° C. until the reaction becomes slow and practically ceases, e. g. for about 20 hours. The condensation can be effected at temperatures as low as 150° C. and as high as 250° C. without great difficulty. However, at the lower temperatures longer reaction times (20 to 30 hours) are necessary, while at higher temperatures, shorter reaction times (12 to 20 hours) suffice. Temperatures much higher than 250° C. are in general unsatisfactory and for that reason precautions to avoid local heating should be taken. This can be done by agitating the reaction mass. In a closed system, the pressure rises to about 225 pounds per square inch at 200° C., when amyl amines are used and to somewhat higher pressures when butylamines are used.

During the reaction, water is formed. This water of formation and any excess of amine can be distilled off following completion of the reaction. The amine thus recovered can be separated from the water which also distils by any suitable means. We have found that certain amines form azeotropes with water and hence cannot be separated from the water by fractional distillation. Amylamines are particularly difficult to handle in this respect. A satisfactory method of separating the amines from water is to add a polyhydric phenol to the amine-water mixture, when the amine and polyhydric phenol form a salt. The water can then be removed from this salt by introducing a liquid which forms an azeotrope with water boiling lower than the decomposition temperature of the salt. Diisopropyl ether is suitable for this purpose, in the case of amylamines and hydroquinone.

We can subject our new reaction products to distillation under reduced pressure in order to effect a purification, if desired. Pressures of 2 mm. of mercury or less are suitable. So distilled, our products are pale yellow liquids which solidify slowly on standing. The distillation serves to remove suspended carbonaceous and tarry matter but does not serve to separate our reaction products into discreet substances.

The following examples serve to illustrate but are not intended to limit our invention:

Example 1.—275 lbs. (1.25 mols) of primary normal butyl primary amine (containing about 9% by weight of water and other inert materials) were changed into a still provided with an agitator, a simple fractionating column, steam and cooling coils, surface condenser and an automatic liquid separator. 110 lbs. (1 mol) of hydroquinone were added and the whole was agitated until solution of the hydroquinone was effected. Heat may be applied to effect this solution. This so prepared solution was conveyed to an autoclave and heated therein at about 200° C. for about 20 hours. Following this treatment the reaction mixture was subjected to distillation in a rapid circulating evaporator, when any excess butylamine and the water of formation distilled off. The residue was thoroughly dried and degassed of remaining traces of butylamine. The degassed material was transferred to a high vacuum still and distilled under a pressure of 2 mm. or less. The product is a pale yellow liquid which solidifies slowly on standing. The product needs no further purification. It can be used as a motor fuel stabilizer in the solid form, but is preferably introduced into the motor fuel from a 50% (weight) solution in a solvent composed of 90% methanol and 10% butanol or other suitable solvent.

Example 2.—557 lbs. (6.0 mols) of commercial primaryamylprimaryamine (usually containing about 10% by weight of water and other inert materials, usually having a boiling range from about 85° C. to about 105° C. and comprising a plurality of isomeric amines) were charged into a still provided with an agitator, a simple fractionating column, steam and cooling coils, surface condenser and an automatic liquid separator. 300 lbs. (2.7 mols) of hydroquinone were added and the whole was agitated until solution of the hydroquinone was effected. Heat may be applied to effect this solution. This so prepared solution was conveyed to an autoclave and heated therein at about 200° C. for about 20 hours. Following this treatment the reaction mixture was subjected to distillation in a rapid circulating evaporator, when the excess amylamine and the water of formation distilled off. The residue was thoroughly dried and degassed of remaining traces of amylamine. The degassed material was transferred to a high vacuum still and distilled under a pressure of about 2 mm. or less. The product is a pale yellow liquid which solidifies slowly on standing. The product needs no further purification. It can be used as a motor fuel stabilizer in the solid form, but is preferably introduced into the motor fuel from a 50% (weight) solution in a solvent composed of 90% methanol and 10% butanol or other suitable solvent.

The excess amylamines and the water of formation which distilled off from the rapid circulating evaporator were returned to the mixing tank. 100 gallons of diisopropyl ether and 300 lbs. of hydroquinone were added. The whole was distilled at atmospheric pressure. The water and diisopropyl ether distilled as an azeotrope leaving a residue which was a salt of hydroquinone and amylamines. This residue can be used in preparing the next batch of reaction product. Instead of diisopropyl ether any liquid can be used, which forms a suitable azeotrope with water, i. e., one boiling lower than the decomposition temperature of the phenol-amine salt. Any alkyl amine forming a salt with a polyhydric phenol can be separated from water in this manner.

The better products for the stabilization of motor fuels, prepared as indicated above, appear to contain a weight of attendant reaction products equal to from about 5% to about 25% of the total weight of both the principal and attendant reaction products. Smaller amounts of these attendant reaction products have a beneficial effect and products containing 50% by weight or more of attendant reaction products are useful as motor fuel stabilizers.

The polyhydric phenols employed can be substituted on the nucleus by groups such as methyl, methoxy and the like. Catechol, hydroquinone and pyrogallol yield products which are powerful motor fuel stabilizers, but from all standpoints these products prepared as indicated above, from either unsubstituted catechol or hydroquinone, particularly the latter, are especially useful as motor fuel stabilizers. About two and one-half molecular proportions of amine for each molecular proportion of pyrogallol is ordinarily most advantageous.

Various higher primary or secondary alkyl primary amines, such as normal primary butyl, normal secondary butyl, normal primary amyl, isoamyl, isohexyl, isobutyl and the like as well as mixtures thereof can be used. The secondary alkyl amines appear to react more slowly than the primary alkyl amines and accordingly the higher of the reaction temperatures given above are ordinarily advantageous when employing secondary alkyl amines. Frequently commercial amines contain mixtures of primary and secondary amines and because of the slower reaction of the secondary amines they will ordinary be found to accumulate in the recovered amines. The reaction products prepared from the primary alkyl primary amines are definitely superior to those prepared from secondary alkyl primary amines as motor fuel stabilizers.

In preparing our new reaction products, as indicated above, catalysts can be added to the reaction mixtures or the amines can be used in the form of derivatives which decompose under the conditions of the reaction to yield the amine.

In accordance with accepted chemical nomenclature, "primary alkyl" is intended to mean an alkyl group derived from a primary alcohol by dropping the OH group; "secondary alkyl" to means an alkyl group derived from a secondary alcohol by dropping the OH group; "N-alkyl", "N-methyl" and "N-benzyl" to mean that the alkyl, methyl or benzyl group is attached to nitrogen.

What we claim as our invention and desire to be secured by Letters Patent of the United States of America is:

1. An aminophenol selected from the group consisting of N-alkyl-o-aminophenols and N-alkyl-p-aminophenols in which the alkyl group contains at least four carbon atoms, together with the attendant reaction products formed when preparing the N-alkylaminophenol from a primary amine selected from the group consisting of primary alkyl and secondary alkyl primary amines, having the general formula $C_nH_{2n+1}NH_2$, wherein n represents a positive integer not smaller than four, and a polyhydric phenol selected from the group consisting of polyhydric phenols of the benzene series containing at least two phenolic hydroxyl groups in ortho position to one another and polyhydric phenols of the benzene series containing at least two phenolic hydroxyl groups in para position to one another.

2. A N-alkyl-p-aminophenol in which the alkyl group contains at least four carbon atoms together with the attendant reaction products formed when preparing the N-alkyl-p-aminophenol from hydroquinone and a primary alkyl primary amine of the following fomula

$$C_nH_{2n+1}NH_2$$

wherein n represents a positive integer not smaller than four.

3. A N-butyl-p-aminophenol together with the attendant reaction products formed when preparing the N-butyl-p-aminophenol from hydroquinone and a primary butyl primary amine having the formula $C_4H_9NH_2$.

4. A N-amyl-p-aminophenol together with the attendant reaction products formed when preparing the N-amyl-p-aminophenol from hydroquinone and a primary amyl primary amine having the formula $C_5H_{11}NH_2$.

5. A process for preparing an aminophenol composition, useful as a motor fuel stabilizer, comprising reacting with the elimination of water, a polyhydric phenol selected from the group consisting of polyhydric phenols of the benzene series containing at least two phenolic hydroxyl groups in ortho position to one another and polyhydric phenols of the benzene series containing at least two phenolic hydroxyl groups in para position to one another with a primary amine selected from the group consisting of primary and secondary alkyl primary amines having the general formula $C_nH_{2n+1}NH_2$ wherein n represents a positive integer not smaller than four.

6. A process for preparing an aminophenol composition, useful as a motor fuel stabilizer, comprising reacting with the elimination of water, in the presence of a diluent, one molecular proportion of a polyhydric phenol selected from the group consisting of polyhydric phenols of the benzene series containing at least two phenolic hydroxyl groups in ortho position to one another and polyhydric phenols of the benzene series containing at least two phenolic hydroxyl groups in para position to one another with from about one and a quarter molecular proportions to about two and a half molecular proportions of a primary amine selected from the group consisting of primary and secondary alkyl primary amines having the general formula $C_nH_{2n+1}NH_2$ wherein n represents a positive integer not smaller than four, the amount of said diluent equaling not more than about ten percent by weight of the primary amine.

7. A process for preparing a composition of matter, useful as a motor fuel stabilizer, comprising reacting, at a temperature of from about 150° C. to about 250° C., one molecular proportion of a polyhydric phenol selected from the group consisting of polyhydric phenols of the benzene series containing at least two phenolic hydroxyl groups in ortho position to one another and polyhydric phenols of the benzene series containing at least two phenolic hydroxyl groups in para position to one another with from about one and a quarter to about two and a half molecular proportions of a primary amine selected from the group consisting of primary and secondary alkyl primary amines having the general formula $C_nH_{2n+1}NH_2$ wherein n represents a positive integer not smaller than four.

8. A process for preparing a composition of matter, useful as a motor fuel stabilizer, comprising reacting, at a temperature of from about 150° C. to about 250° C. and in a closed vessel, one molecular proportion of a polyhydric phenol selected from the group consisting of polyhydric phenols of the benzene series containing at least two phenolic hydroxyl groups in ortho position to one another and polyhydric phenols of the benzene series containing at least two phenolic hydroxyl groups in para position to one another with from about one and a quarter to about two and a half molecular proportions of a primary amine selected from the group consisting of primary and secondary alkyl primary amines having the general formula $C_nH_{2n+1}NH_2$ wherein n represents a positive integer not smaller than four.

9. A process for preparing a composition of matter, useful as a motor fuel stabilizer, comprising reacting, at a temperature of from about 150° C. to about 250° C. in a closed vessel and in the presence of a diluent, one molecular proportion of a polyhydric phenol selected from the group consisting of polyhydric phenols of the benzene series containing at least two phenolic hydroxyl groups in ortho position to one another and polyhydric phenols of the benzene series containing at least two phenolic hydroxyl

groups in para position to one another with from about one and a quarter to about two and a half molecular proportions of a primary amine selected from the group consisting of primary alkyl and secondary alkyl primary amines having the general formula $C_nH_{2n+1}NH_2$ wherein n represents a positive integer not smaller than four, the amount of said diluent equaling not more than about ten percent by weight of the primary amine.

10. A process for preparing a composition of matter, useful as a motor fuel stabilizer, comprising reacting, at a temperature of from about 150° C. to about 250° C. in a closed vessel and in the presence of a diluent, one molecular proportion of a polyhydric phenol selected from the group consisting of polyhydric phenols of the benzene series containing at least two phenolic hydroxyl groups in ortho position to one another and polyhydric phenols of the benzene series containing at least two phenolic hydroxyl groups in para position to one another with from about one and a quarter to about two and a half molecular proportions of a primary alkyl primary amine having the general formula $C_nH_{2n+1}NH_2$ wherein n represents a positive integer not smaller than four, the amount of said diluent equaling not more than about ten percent by weight of the primary amine.

11. A process for preparing a composition of matter, useful as a motor fuel stabilizer, comprising reacting, at a temperature of from about 150° C. to about 250° C. and in a closed vessel, one molecular proportion of hydroquinone with from about one and a quarter to about two and a half molecular proportions of a primary amine selected from the group consisting of primary and secondary alkyl primary amines having the general formula $C_nH_{2n+1}NH_2$ wherein n represents a positive integer not smaller than four.

12. A process for preparing a composition of matter, useful as a motor fuel stabilizer, comprising reacting, at a temperature of from about 150° C. to about 250° C., one molecular proportion of hydroquinone with from about one and a quarter to about two and a half molecular proportions of a primary alkyl primary amine having the general formula $C_nH_{2n+1}NH_2$ wherein n represents a positive integer not smaller than four.

13. A process for preparing a composition of matter, useful as a motor fuel stabilizer, comprising reacting, at a temperature of from about 150° C. to about 250° C. and in a closed vessel, one molecular proportion of hydroquinone with from about one and a quarter to about two and a half molecular proportions of a primary alkyl primary amine having the general formula $C_nH_{2n+1}NH_2$ wherein n represents a positive integer not smaller than four.

14. A process for preparing a composition of matter, useful as a motor fuel stabilizer, comprising reacting, at a temperature of from about 150° C. to about 250° C. in a closed vessel and in the presence of a diluent, one molecular proportion of hydroquinone with from about one and a quarter to about two and a half molecular proportions of a primary alkyl primary amine having the general formula $C_nH_{2n+1}NH_2$ wherein n represents a positive integer not smaller than four, the amount of said diluent equaling not more than about 10% by weight of the primary amine.

15. A process for preparing a composition of matter, useful as a motor fuel stabilizer, comprising reacting, at a temperature of from about 150° C. to about 250° C. in a closed vessel and in the presence of a diluent, one molecular proportion of hydroquinone with from about one and a quarter to about two and a half molecular proportions of a primary butyl primary amine having the formula $C_4H_9NH_2$, the amount of said diluent equaling not more than about 10% by weight of the primary amine.

16. A process for preparing a composition of matter, useful as a motor fuel stabilizer, comprising reacting, at a temperature of from about 150° C. to about 250° C. in a closed vessel and in the presence of a diluent, one molecular proportion of hydroquinone with from about one and a quarter to about two and a half molecular proportions of a primary amyl primary amine having the formula $C_5H_{11}NH_2$, the amount of said diluent equaling not more than about 10% by weight of the primary amine.

17. A process for preparing a composition of matter, useful as a motor fuel stabilizer, comprising reacting, at a temperature of about 200° C. in a closed vessel and in the presence of a diluent, one molecular proportion of hydroquinone with from about one and a quarter to about two and a half molecular proportions of a primary alkyl primary amine having the general formula $C_nH_{2n+1}NH_2$ wherein n represents a positive integer not smaller than four, the amount of said diluent equaling not more than about 10% by weight of the primary amine.

18. A process for preparing a composition of matter, useful as a motor fuel stabilizer, comprising reacting with the elimination of water, at a temperature of from about 150° C. to about 250° C. in a closed vessel, one molecular proportion of a polyhydric phenol selected from the group consisting of polyhydric phenols of the benzene series containing at least two phenolic hydroxyl groups in ortho position to one another and polyhydric phenols of the benzene series containing at least two phenolic hydroxyl groups in para position to one another with from about one and a quarter to about two and a half molecular proportions of a primary amine selected from the group consisting of primary alkyl and secondary alkyl primary amines having the general formula $C_nH_{2n+1}NH_2$ wherein n represents a positive integer not smaller than four, separating the unreacted primary amine and the water, admixing a polyhydric phenol with the primary amine and water to form a salt between the polyhydric phenol and the primary amine, admixing diisopropyl ether with the salt and water and distilling to remove the water and ether as an azeotrope.

19. In a process which gives rise to a mixture of water and an alkyl amine, a process for separating the water and the alkyl amine comprising admixing a polyhydric phenol with the alkyl amine and water to form a salt between the alkyl amine and polyhydric phenol, admixing diisopropyl ether with the mixture of the salt and water and distilling to remove the water and ether as an azeotrope.

HAROLD VON BRAMER.
ALBERT C. RUGGLES.

Certificate of Correction

Patent No. 2,163,639. June 27, 1939.

HAROLD VON BRAMER ET AL.

It is hereby certified that errors appear in the printed specification of the above numbered patent requiring correction as follows: Page 3, first column, line 55, for "fomula" read *formula;* and second column, line 62, for "$C_nH\ _{n+1}NH_2$" read $C_nH_{2n+1}NH_2$; and that the said Letters Patent should be read with these corrections therein that the same may conform to the record of the case in the Patent Office.

Signed and sealed this 19th day of September, A. D. 1939.

[SEAL]

HENRY VAN ARSDALE,
Acting Commissioner of Patents.

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BRIEF FOR THE COMMISSIONER OF PATENTS, APPELLEE

In the United States Court of Appeals for the District of Columbia

No. 8170

Eastman Kodak Company, appellant
v.
Conway P. Coe, Commissioner of Patents, appellee

APPEAL FROM THE JUDGMENT OF THE DISTRICT COURT OF THE UNITED STATES FOR THE DISTRICT OF COLUMBIA

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In the United States Court of Appeals for the District of Columbia

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APPEAL FROM THE JUDGMENT OF THE DISTRICT COURT OF THE UNITED STATES FOR THE DISTRICT OF COLUMBIA

BRIEF FOR THE COMMISSIONER OF PATENTS

INTRODUCTION

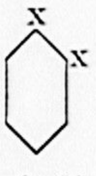
This is an appeal from the judgment (Appellant's App. 8) of the District Court of the United States for the District of Columbia dismissing the complaint of appellant, the Eastman Kodak Company, which complaint was brought under Section 4915 R. S. (U. S. C., title 35, section 63) and sought to have said court authorize the Commissioner of Patents to issue to appellant, as assignee, a patent containing claims 28 and 29 of an application, Serial No. 134,903, filed by Frederic R. Bean, in the United States Patent Office on April 3, 1937. These appealed claims are found in the complaint (Appellant's App. 2).

THE BEAN APPLICATION

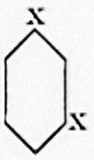
The specification (Appellant's App. 65) of the Bean application relates this applicant's alleged contribution to the chemical art to the broad field of N-substituted aminophenols, meaning nitrogen substituted aminophenols, and within that field, particularly to N-alkyl aminophenols, including N-ethyl-p-aminophenol, N-(n-primarybutyl)-p-aminophenol, N-(n-primaryamyl)-p-aminophenol, and N-(2-ethyl-n-primarybutyl)-p-aminophenol (Appellant's App. 67, 69). The specification includes descriptions of these compounds and of a process for producing them. The specification emphasizes the utility of the compounds as agents to retard the deterioration of motor fuels. Claims to the process have been allowed (Appellant's App. 108–109). It thus has been recognized that the process is new and useful, and that it is an invention.

No claims covering any of the chemical compounds so disclosed have been allowed, either by the Patent Office or the District Court. As appears from original claims 1 to 6 (Appellant's App. 72) and from the claims here on appeal (Appellant's App. 2), Bean has never sought protection on the compound, N-(n-primaryamyl)-p-aminophenol. However, before the Court of Customs and Patent Appeals, two joint applicants, Von Bramer and Ruggles, did attempt to obtain claims to a motor fuel containing that compound, and, as indicated by the reported decision of that court (—— C. C. P. A. (Patents) ——, 127 F. (2d) 149), were unsuccessful.

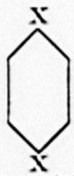
Of the two claims on appeal, claim 29 is a specific claim, a claim for one particular chemical compound. In accordance with the specification (Appellant's App. 68), which sets forth the same formula as that set forth in claim 29, that compound is most accurately described as N-(n-primarybutyl)-p-aminophenol, meaning Nitrogen-(normal-primarybutyl)-para-aminophenol. Other accurate names will be referred to hereinafter. An "aminophenol" is (Appellant's App. 37) an aromatic compound and hence is derived from the hydrocarbon benzene, C_6H_6, represented in the formula by the hexagon; for one hydrogen atom of the benzene there has been substituted the "hydroxyl" group, OH, by which substitution benzene becomes "phenol," and for still another hydrogen atom there has been substituted the "amino" group, NH_2, by which such further substitution, the "phenol" becomes "aminophenol." An "alky aminophenol" is an aminophenol in which for one hydrogen atom of the amino group, NH_2, or for still another hydrogen atom of the benzene, there has been substituted an "alkyl radical" having the general formula C_nH_{2n+1}, where "n" represents an integer. An "N-substituted aminophenol" or an "N-alkyl aminophenol" is an aminophenol in which the substitution of the alkyl radical is in the amino group, NH_2. Where, in such case, the alkyl radical which is substituted is the "butyl" radical (C_4H_9—or, the equivalent in the formula, CH_3—CH_2—CH_2—CH_2—, and n=4), then the "N-aminophenol" is "N-butyl aminophenol." The amino group, together with the substituting butyl radical, C_4H_9, represented in the formula as $CH_3—CH_2—CH_2—CH_2—NH—$, and the hydroxyl group, OH, may be substituted in the benzene ring for two hydrogen atoms thereof in accordance with the following scheme, to form disubstitution products designated by the stated names and abbreviations:



ortho (o)



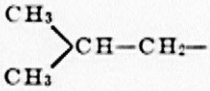
meta (m)

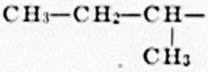


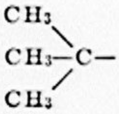
para (p)

Thus, the formula set forth in claim 29 represents "N-butyl-para-aminophenol."

There are four possible forms of the butyl radical. Said formula of claim 29 sets forth the "normal" or straight chain butyl radical. "Normal" is abbreviated as "n." The entire formula thus represents "N- (n-butyl) -p-aminophenol." The three other possible forms of the butyl radical are:

II. Isobutyl:

$$\begin{matrix} CH_3 \\ & \rangle CH—CH_2— \\ CH_3 \end{matrix}$$

III. Secondarybutyl:

$$CH_3—CH_2—\underset{\displaystyle CH_3}{\underset{|}{C}}H—$$

IV. Tertiarybutyl:

$$\begin{matrix} CH_3 \\ & \diagdown \\ CH_3— & C— \\ & \diagup \\ CH_3 \end{matrix}$$

Because, unlike the foregoing butyl radicals III and IV, the combining carbon atom of the normal butyl radical set forth in the formula of claim 29 (which is the first carbon atom to the left of the benzene ring, or hexagon) is linked to only one other carbon atom in that straight chain, the butyl radical is not only properly designated "n-butyl" but also "n-primarybutyl." Thus, with extreme scientific exactitude, the formula of claim 29 represents "N-(n-primarybutyl)-p-aminophenol," which exact terminology, as pointed out later herein, is largely confined to Bean's specification. Since "normal," by definition, is restricted to a straight chain arrangement, and "primary," by definition, requires that the combining carbon atom be at the end of a chain and linked to only one other carbon atom in the chain, as in the butyl radical of claim 29, or as in the isobutyl radical of II, above, the word "primary" is actually unnecessary to describe the compound of claim 29 and to modify "normal," since a "normal" butyl radical must be "primary" as well.

Claim 28 is a claim of the so-called "Markush" type (*Ex parte Markush*, 340 O. G. 839, 1925 C. D. 126). The general formula set forth in this claim represents three different compounds. Where "R" is hydrogen, the compound is the specific compound represented by the formula of claim 29, to wit: "N-(n-primarybutyl)-p-aminophenol." Where "R" is the ethyl radical, an alkyl radical in which the integer "n" is 2, then the compound is "N-(2-ethyl-n-primarybutyl)-p-aminophenol," whose formula and description are given in the Bean specification (Appellant's App. 69–70). "R" may also be the methyl radical (n=1), to give the compound "N-(2-methyl-n-primarybutyl)-p-aminophenol." The numeral "2" refers to the position of "R" in the straight chain butyl radical in accordance with the accepted scheme of chemical nomenclature. There are four positions as follows:

$$\underset{4}{CH_3}—\underset{3}{CH_2}—\underset{2}{CH_2}—\underset{1}{CH_2}—$$

Since claim 28 is, in effect, a broad claim, any prior art that anticipates claim 29 renders claim 28 unpatentable (*In re Steenbock,* 23 C. C. P. A. (Patents) 1244, 83 F. (2d) 912).

THE PRIOR ART RELIED UPON BY APPELLEE

While six patents were cited as references by the Examiner in his statement (Appellant's App. 108) and the same six patents were set up in the answer (Appellant's App. 4), only three of these six are now relied upon on this appeal. They are the United States patent to Clifford, No. 2,109,164 (Appellant's App. 99), granted February 22, 1938, upon an application filed October 22, 1932; the British patent to Wade, No. 350,438 (Appellant's App. 103), granted June 5, 1931; and the United States patent to Rogers and Voorhees, No. 2,084,977 (Appellant's App. 98), granted June 22, 1937, upon an application filed March 26, 1937, as a "continuation-in-part" of a parent application filed February 10, 1930 (Appellant's App. 86). Since Bean's dates of invention (Finding of Fact No. 7, Appellant's App. 7) are prior to the filing date of the Rogers and Voorhees continuation-in-part application, though not prior to the filing date of said parent application, it is conceded that the Rogers and Voorhees patent is a reference against Bean upon the claims in issue only as to matter commonly disclosed therein and in the parent application.

The Rogers and Voorhees patent speaks as an anticipation as of its effective filing date, to wit: February 10, 1930, and the Clifford patent as of its filing date, to wit: October 22, 1932 (*Detrola Radio and Television Corporation* v. *Hazeltine Corporation,* 313 U. S. 259; *Alexander Milburn Co.* v. *Davis-Bournonville Co.,* 270 U. S. 390).

The Clifford patent (Appellant's App. 99, column 1, lines 1 to 10) expressly relates to the retarding of the ageing of deteriorable organic compositions, such as rubber and gasoline, by the incorporation therein of age retarders or antioxidants. In the case of gasoline, Clifford asserts (Appellant's App. 99, column 1, lines 15 to 17) that the antioxidants prevent the formation of gums and gummy materials. Clifford specifies (Appellant's App. 100, column 2, line 8) "N-butyl p-amino phenol" as one such age retarder or antioxidant. He suggests (Appellant's App. 101, column 2, lines 18 to 20) a method of preparing that compound. Most importantly, Clifford (Appellant's App. 102, column 2, lines 1 to 15) gives data on tests of rubber samples in which was incorporated "N-butyl p-amino phenol."

The British patent to Wade (Appellant's App. 103) relates to improvements in hydrocarbon motor fuel products, and, specifically, in gasoline (Appellant's App. 103, column 1, lines 13–15). The need for improvement is discussed at length in following sections of the patent. The improvement consists in treating gasoline with a "substituted aminophenol" (Appellant's App. 104, column 2, lines 85 to 91).

Wade states (Appellant's App. 104, column 2, lines 100 to 105):

> As examples of such materials, we have employed *para-* and ortho-methyl-amino-phenol, dimethyl-amino-phenol, *butyl,* isopropyl and benzyl *amino-phenols,* dihydroxy-diphenylamine, and the like. [Italics added.]

The primary examiner (Appellant's App. 109), the Board of Appeals (Appellant's App. 113), and the District Court (Appellant's App. 6, Finding of Fact No. 4) have all construed this quotation as a disclosure of para-butyl-aminophenol.

Properties of these substituted aminophenols and the gasoline incorporating them are discussed at length (Appellant's App. 104 and 105) with reference to anti-knock values, gum formation, induction period, reaction to dilute alkalies and acids, etc. Wade asserts (Appellant's App. 105, column 2, lines 97 to 104) that his substituted aminophenols are much more effective than the aminophenols.

The Rogers and Voorhees parent application (Appellant's App. 86) contains a disclosure of para-butyl-aminophenol and its use as an inhibitor in gasoline in a manner not differing in substance from the disclosure in the Wade British patent (Appellant's App. 103), and so the Rogers and Voorhees patent (Appellant's App. 98) and said Wade patent may be considered to be about on a par as references.

COUNTER STATEMENT OF THE CASE

The suit under Section 4915 R. S. here on appeal is no different from the usual suit under that section, unless a difference arises from the circumstance that it and another such suit, Civil Action No. 7608, also entitled *Eastman Kodak Company* v. *Coe,* were tried (Appellant's App. 4) and decided together. The findings and conclusions in that other civil action are reported in 51 U. S. P. Q. 407. That civil action involved an application of Bean, Serial No. 175,078, as a "division" of the application involved in this appeal. That application of Bean, Serial No. 175,078, is several times referred to in the decision of the Court of Customs and Patent Appeals in *In re Von Bramer and Ruggles, supra.*

As below and in the Patent Office, the main issue on this appeal is whether the appealed claims 28 and 29 are met by the cited references. On behalf of the Commissioner of Patents, it is maintained that claim 29 is completely anticipated by the patents relied upon and that claim 28 is not patentable over these patents.

SUMMARY OF ARGUMENT

1. A chemical compound is proper subject matter for a United States patent. However, appellant's compound, N-(n-primarybutyl)-p-aminophenol, is recited in the three patents upon which the Commissioner relies. These patents bear dates earlier than the date when Bean, appellant's assignor, invented that compound. Hence, these patents anticipate claims 28 and 29 on appeal, which generically and specifically define that compound.

2. The prior art patents do more than list the "mere" names of chemical compounds. These patents recite properties and show tests of the claimed

compound. The compound was not "unknown." In scientific nomenclature, there is no such thing as a "mere" name.

3. Appellant does not assert what description is necessary in a reference patent to anticipate a claimed chemical compound. Under the well-settled law, disclosure of a method of preparing the compound is not necessary. The District Court therefore correctly held (Conclusion of Law No. 3, Appellant's App. 8) that the prior art patents anticipate appellant's claimed compound, regardless of whether they do or do not disclose a process for preparing the compound, and regardless of whether or not they disclose a process for preparing the compound which is operative. The cited Clifford patent does disclose one presumptively operative method of preparing that compound.

4. There is intrinsic evidence in the cited prior art that the compound disclosed therein and defined by the claims on appeal did "exist" earlier than Bean's date of invention. Evidence for appellant does not establish the contrary.

5. Appellant's claimed compound is described in definite terms in the prior art.

6. Claim 29 is anticipated by the prior art, and, by the same token, claim 28 is unpatentable thereover. Conclusion of Law No. 1 (Appellant's App. 8) is correct. Finding of Fact No. 8 (Appellant's App. 7) is a safeguarding finding, and is not necessary to support said conclusion. Said finding is amply supported by the evidence.

7. There is no "conflict of decisions."

ARGUMENT

Appellant's chemical compound is anticipated

1. It is not denied that a chemical compound is proper subject matter for a United States patent, though the law is different in several foreign countries.[1] However, it is maintained that to receive such patent protection, the compound must be new, useful, and an invention (*The Cuno Engineering Corporation* v. *The Automatic Devices Corporation*, 314 U. S. 84; *Thompson* v. *Boisselier*, 114 U. S. 1). As already stated, claims 28 and 29 here on appeal define generically and specifically the compound, N-(n-primarybutyl)-p-aminophenol. That compound is not entitled to patent protection because the same compound was invented by others, Rogers and Voorhees, and Clifford, prior to Bean (*Alexander Milburn Co.* v. *Davis-Bournonville Co., supra; Detrola Radio and Television Corporation* v. *Hazeltine Corporation, supra*). It was also patented and described by another, Wade, in a printed publication prior to Bean's date of invention (Section 4886 R. S., U. S. C., title 35, section 31).

Nomenclature

2. Appellant complains that the three patents upon which the Commissioner here relies do no more than list the mere names of allegedly unknown chemical compounds. From the discussion of these patents hereinabove, it is plain that this complaint is not supportable. Properties by which the compounds may be

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[1] Lewers, Composition of Matter (1922) 4 J. Pat. Off. Soc. 530; Federico, Patents for New Chemical Compounds (1939) 21 J. Pat. Off. Soc. 544.

distinguished are recited in each patent. And certainly it is not to be presumed that Clifford secured the test data recorded on page 4 of his patent (Appellant's App. 102, column 2, lines 5 to 15) from the mere theoretical consideration of an imagined compound, "N-butyl p-amino phenol." The compound so designated and the same compound described in the Wade (British) and Rogers and Voorhees patents as "para-butyl-aminophenol," was not unknown prior to Bean's date of invention thereof. Actual knowledge was imparted by the Wade (British) patent as of the date it was granted. The law presumes the impartation of knowledge by the Clifford, and Rogers and Voorhees patents as of, respectively, the filing date and effective filing date thereof (*Alexander Milburn Co.* v. *Davis-Bournonville Co., supra; Detrola Radio and Television Corporation* v. *Hazeltine Corporation, supra*).

In biological and chemical nomenclature there is no such thing as a mere name. Thus, in the field of botany, the rose commonly known as "Austrian Copper" is scientifically termed "Rosa Foetida Bicolor," thus distinguishing that rose from other roses as smelling to heaven and also as having two distinct colors. The subject of chemical nomenclature is extensively dealt with in the decision of the Court of Customs and Patent Appeals in *In re Von Bramer and Ruggles, supra.* There the contention also was raised that disclosure in the prior art of a claimed compound no further than by the customary chemical nomenclature was insufficient to anticipate the appellants' claims. In turning down this contention, the Court quoted with approval the following statement of the examiner having reference to the very Clifford patent relied upon by the Commissioner in the present appeal:

> the N-butyl amino phenol mentioned by the patentee is more than a mere name of an individual substance, otherwise unrecognized. This is a name according to a standard system of chemical nomenclature (Geneva system) whereby a chemically individual substance of definite chemical molecular structure is defined having generally predictable properties such as found for any similar N-alkyl amino phenol, and generally capable of synthesis by the recognized classical organic reactions. The use of a name falling within the standard chemical system by the patentee is no accident because all of the many compounds named in Clifford's disclosure are deliberately named from this standard chemical system.

Dr. Davy admits (Appellant's App. 38–39) that the Rogers and Voorhees parent application (Appellant's App. 90), in its disclosure of "butyl aminophenol" would, by those very words, tell the chemist familiar with aminophenols much of its expected properties; he mentions solubility, chemical reactivity, and color, properties stressed in the Von Bramer and Ruggles appeal to the Court of Customs and Patent Appeals. So here is further proof that a chemical name is no mere name.

Necessary description in a reference patent

3. Von Bramer and Ruggles in their aforesaid appeal to the Court of Customs and Patent Appeals

were more specific than appellant here as to what, in their opinion, was the required description of a chemical compound in a patent depended upon for anticipation. There, Von Bramer and Ruggles contended that a chemical compound can be described in only two ways: (1) by reciting a sufficient number of its physical and chemical attributes as "melting point, boiling point, color, crystalline appearance, solubility, chemical reactivity, etc.," or (2) "to recite a process which will unquestionably produce the substance." This contention found no favor with said Court, and that Court ruled that:

> It is not necessary that a reference patent for a device or chemical compound disclose an operative process for producing the article or product. *In re Marden et al.*, 48 F. (2d) 428, 18 C. C. P. A., Patents, 1119; *In re Fink,* 62 F. 2d 103, 20 C. C. P. A., Patents, 716; *Cohn* v. *United States Corset Co.,* 93 U. S. 366, 23 L. Ed. 907; *In re Appeal of Schaeffer,* 2 App. D. C. 1; *In re Decker,* 36 App. D. C. 104; *General Electric Co.* v. *De Forest Radio Co., et al.,* D. C., 17 F. 2d 90; *One-Piece Bifocal Lens Co.* v. *Bisight Co. et al.,* D. C., 246 F. 450.

The cited case of *In re Schaeffer* (2 App. D. C. 1) is directly in point here. In that case, this Court said:

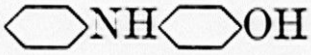
> In this connection it must be remembered that appellant's claim for a new invention does not consist in the method of rendering alizarin soluble in water, nor in a new process of dyeing, but exclusively in the mixture itself of alizarin, boracic acid, and an alkali.
>
> It is well settled that where the claim is for a new manufacture, and not for the process of

> making it, it is not required that the prior publication shall be so specific as to teach how to make the article.
>
> The requirement is that the thing sought to be patented shall have been described, and not the necessary steps antecedent to its production (*Cohn* v. *U. S. Corset Co.*, C. D., 1877, 205; 11 O. G. 457; 93 U. S. 366).

This quoted rule, and the rule of the other cases cited by the Court of Customs and Patent Appeals, arises from the language of Section 4886 R. S. (U. S. C., title 35, section 31), which requires merely a description in a printed publication of a composition of matter to invalidate a patent upon the composition of matter. On that score, that section differs from Section 4888 R. S. (U. S. C., title 35, section 33), which requires the applicant for the patent to file a written description of his composition of matter and "of the manner and process of making, constructing, compounding, and using it."

Thus, the District Court correctly ruled (Appellant's App. 8), as a matter of law, that disclosure of the compound N-(n-primarybutyl)-p-aminophenol in the cited prior art is an anticipation of that compound, regardless of disclosure of any process for preparing that compound.

In view of the established rule of law that a reference patent for a chemical compound need not disclose an operative process for producing that compound, the evidence offered by appellant, particularly the testimony of Vaughn (Appellant's App. 33–34) and Dr. Davy (Appellant's App. 45 to 48), to show that the method suggested by Clifford (Appellant's App. 101, column 2, lines 18 to 20) for producing N-butyl-p-aminophenol is inoperative, could well be disregarded as immaterial, and appellant's contention of inoperativeness might well be ignored. However, for the benefit of this Court, it will be pointed out that Clifford does suggest (Appellant's App. 100, column 2, lines 60 to 75) a method which presumptively would be operative to produce N-butyl-p-aminophenol and which appellant has not proved is inoperative for that purpose. The method is set forth in connection with the preparation of para hydroxy diphenyl amine, but it has more general application. Para hydroxy diphenyl amine (which may also be identified as N-phenyl-p-aminophenol) is a compound having the following formula:



To prepare that compound, Clifford adds hydroquinone to aniline. Aniline is an aromatic amine (Appellant's App. 23), to wit: phenylamine, having the formula NH$_2$. The use of aniline, an aromatic amine, would suggest the use of butylamine, an aliphatic amine, to prepare, not para hydroxy diphenyl amine, but N-butyl-p-aminophenol. The significance of the Von Bramer and Ruggles patent (Appellant's App. 116), to which appellant so strongly objects, is not as prior art, for its filing date is later than the date when Bean first prepared N-(n-primarybutyl)-p-aminophenol, but as proof of the force of this suggestion. Under conditions similar to those recited by Clifford (Appellant's App. 100, column 2, lines 60 to 75) for the preparation of para hydroxy diphenyl amine by adding hydroquinone to aniline, Von Bramer and Ruggles prepared (Appellant's App. 117, column 1, lines 49 to 71) what is N-(n-primarybutyl)-p-aminophenol (Appellant's App. 22, 23, 57) by adding hydroquinone to butylamine. This Von Bramer and Ruggles patent indicates that for appellant to prove that Clifford does not teach any operative process for producing N-butyl-p-aminophenol, it was necessary to try both the method suggested (Appellant's App. 101, column 2, lines 18 to 20) for preparing that compound and the method suggested (Appellant's App. 100, column 2, lines 60 to 75) for preparing para hydroxy diphenyl amine.

Appellant's compound did earlier "exist"

4. The point has hereinbefore been made that there is no such thing as a *mere* chemical name, that a chemical name conveys much information as to properties and suggests processes of preparation. The point has also been made that disclosure of a process of preparation is not necessary in a reference describing a chemical compound to render that reference an anticipation. With these points made, there can now be considered the contention of appellant that there is not "one scintilla of proof" that the "N-butyl-p-amino phenol", of Clifford, or the "butyl aminophenol" of Rogers and Voorhees, and Wade, *existed* at the times when Clifford, and Rogers and Voorhees filed their domestic applications and when Wade received his British patent. Appellant belabors this contention to bring this case within the ruling of Justice Bailey in ***Eastman Kodak Co.*** v. ***Coe,*** 50 U. S. P. Q. 43 and 645, 40 F. Supp. 891.

The description in the several patents to Clifford, Rogers and Voorhees, and Wade of the properties of the compounds therein disclosed has already been mentioned. There have also been mentioned the test data concerning "N-butyl p-amino phenol" recorded in the Clifford patent (Appellant's App. 102, column 2, lines 1 to 15). There may here further be mentioned the statement in the Wade (British) patent (Appellant's App. 104, column 2, lines 100 to 105) that "* * * we have employed para- * * * butyl * * * aminophenols." All this is convincing evidence that the compound did exist. Because of such evidence, it is perhaps not needful to press the contention that, with the compound N-(n-primary-butyl)-p-aminophenol described in chemical terms in the prior art, the burden is upon appellant to show that that compound did not exist before Bean's discovery, and that such a burden cannot be discharged by testimony of the character quoted and referred to by appellant on pages 14 to 18 of its brief. The testimony of Bean, Vaughn and Tanghe in regard to the difficulty of preparing that compound and the alleged failure of the prior art to teach how to prepare the compound is immaterial under the decision of the Court of Customs and Patent Appeals in *In re Von Bramer and Ruggles, supra,* and the authorities cited in that decision. The same, without further comment, holds for much of the testimony of Dr. Davy which is relied upon by appellant. Contrary to appellant's conclusion (Brief, page 18), Dr. Davy does not testify that there was no literature describing the compounds claimed by Bean. His testimony (Appellant's App. 39 and 42) concerns the alleged lack of literature as to preparation, and is as follows:

> Q. Does the name "butyl aminophenol" tell you, speaking as of the date in 1930, how to prepare that compound?
>
> A. Not at all.
>
> Q. In 1930 was there known in the literature any method of preparation of any butyl aminophenol?
>
> A. No.
>
> Q. Did you yourself know any in 1930?
>
> A. No.
>
> Q. Had anyone in 1930 ever told you of any such method?
>
> A. No.
>
> * * * * *
>
> Q. Now, speaking as of 1930, or in fact any time from 1930 to March, 1934, and referring to any literature or art that you are aware of, is there anything about this British patent that tells you how to prepare butyl aminophenol?
>
> A. No; until Bean's latest invention there is nothing in the literature about any method of preparing any butyl aminophenol.

The patents to Rogers and Voorhees, and Clifford here relied upon by the Commissioner were not granted until 1937 and 1938, and yet they, as already mentioned, are available as references against Bean because of their respective effective filing date of 1930 and filing date of 1932. Questioning of Dr. Davy does not even touch upon the possibility of disclosures of methods of preparation in patents granted after

Bean's dates of invention upon applications filed prior thereto.

In view of appellant's reiterated emphasis upon the contribution made by Bean in inventing a process of preparing N-(n-primarybutyl)-p-aminophenol, it must be repeated that the Patent Office has recognized this contribution by allowing claims thereto, and that, once the present suit is terminated, a patent containing such claims may be granted. But the contribution of such method does not entitle Bean to a patent upon the compound when the compound is old.

Appellant's compound is definitely disclosed

5. Appellant offered the testimony of Dr. Davy in an apparent effort to show (Cf. Appellant's App. 37 and 52) that the reference by Clifford in his patent (Appellant's App. 100, column 2, line 8) to "N-butyl p-amino phenol" is a reference to four possible compounds. At least, appellant draws this conclusion (Brief, page 18) from Davy's testimony. Appellant would have it that in the quoted expression the term "butyl" may mean "normal primarybutyl" or "isobutyl" or "secondarybutyl" or "tertiarybutyl" In this, appellant is at odds with the Board of Appeals, which held (Appellant's App. 113):

> * * * if not otherwise described, the simple term "butyl" indicates as universally understood, the simplest form of the butyl radical normal primarybutyl. If iso, secondary, or tertiary forms are intended, these are prefixed to the butyl.

The very Clifford patent indicates such terminology; amongst additional antioxidents Clifford lists (Appellant's App. 100, column 2, line 11) "N-isobutyl p-amino phenol." But perhaps more importantly, as developed on cross-examination, the term "N-butyl-p-aminophenol" is used exclusively in inter-office correspondence between Bean and his colleagues (Appellant's App. 73 to 86) to specify the same compound which appellant claims as, and upon this appeal insistently names "N-(n-primarybutyl)-p-aminophenol." There is even use of the same term by Bean in his testimony (Appellant's App. 13 and 15). Accordingly, it is submitted that the Board's holding is eminently correct, and the District Court's finding (Appellants' App. 6) that "N-butyl p-amino phenol" means N-(n-primarybutyl)-p-aminophenol is likewise correct.

Contrary to the examiner's statement (Appellant's App. 108–109) and the decision of the Board of Appeals (Appellant's App. 113) that the Rogers and Voorhees parent application (Appellant's App. 90, last four lines) and the Wade (British) patent (Appellant's App. 104, column 2, lines 100 to 105) disclose "para-butyl-aminophenol," Dr. Davy asserted (Appellant's App. 36 to 41) that in these cases there is a disclosure only of "butyl aminophenols." So generalizing the disclosures of these cases, Dr. Davy concluded (Appellant's App. 37–38) that 44 different chemical compounds are included under the generic name "butyl aminophenols." Now, apparently the only explanation for so construing the disclosures is that given by Davy under cross-examination (Appellant's App. 49), where, with reference to the Wade

(British), patent, he said, "I think that sticking that 'dimethyl-aminophenol' in there, where you see 'ortho' and 'para' makes all the rest in the general thereafter." This is evidently a grammatical, rather than a chemical explanation, but Dr. Davy did not qualify as a grammarian. It is submitted that the District Court, adopting its own grammatical construction, quite properly found (Appellant's App. 6), in agreement with the examiner and the Board of Appeals, that the parent application of Rogers and Voorhees and the Wade (British) patent do disclose "para-butyl-aminophenol." In discussing the quoted term, the Board (Appellant's App. 113), with reference to claim 2 of Bean's application, Serial No. 175,078, the one involved in Civil Action No. 7608 (51 U. S. P. Q. 407), which claim recited N-(n-primarybutyl)-p-aminophenol as an inhibitor in a motor fuel, said:

> It is the examiner's view that the British patent to Wade No. 350,438 in specifying employment of para-butyl-aminophenol in this relation is a direct anticipation of claim 2 and others in this respect. We are in full agreement with the examiner's holding that unless otherwise specified the above term of the British patent means identically the same compound only more fully described by applicant's term in claim 2. We see no reason for doubt that as to the first point, if not otherwise described, the simple term "butyl" indicates as universally understood, the simplest form of the butyl radical normal primarybutyl. If iso, secondary or tertiary forms are intended, these

> are prefixed to the butyl. We agree further with the examiner's conclusion that unless otherwise specified the term of the British patent also means that the butyl radical is attached to the nitrogen of the amine residue otherwise the term "para" would have no reasonable meaning. If the butyl and amine residues should stand at different positions on the benzene ring with respect to the hydroxyl both positions would have to be specified. It would be unreasonable to assume that the para position might be intended to apply uncertainly to one or the other of the positions of the two substituents.

The District Court (Appellant's App. 6) made like findings. Support for these findings is found in the testimony of Dr. Davy (Appellant's App. 40), and in the use by Bean (Appellant's App. 10) of the term "butyl para aminophenol" as a synonym for "N-(n-"primarybutyl)-p-aminophenol."

Unpatentability of appellant's compounds

6. Since, therefore, Bean's very compound, "N-(n-primarybutyl)-p-aminophenol," as claimed in claim 29 on appeal, is described in each of the patents in the prior art here relied upon by the Commissioner, the District Court correctly held (Appellant's App. 8) that claim 29 is anticipated. The other claim here on appeal, claim 28, is, as already stated, a claim of the "Markush" type (*Ex parte Markush, supra*), and, by that decision, is a broad claim, including within its scope the specific compound claimed in claim 28.

Accordingly, since claim 29 is anticipated by the prior art cited, claim 28 plainly is unpatentable thereover (*In re Steenbock, supra*). The District Court so held (Conclusion of Law No. 1, Appellant's App. 8), and correctly. While these rulings as to anticipation and patentability should have been designated as findings of facts, the error in designating them as conclusions of law was formal and harmless to appellant (*Minnesota Mining and Manufacturing Co.* v. *Coe*, —— App. D. C. ——, 125 F. (2d) 198).

Finding of Fact No. 8 (Appellant's App. 7), as to which appellant also alleges error, is a safeguarding finding. So long as claim 28 is construed, and properly, as a broad claim, and hence to be met by disclosure in the prior art of N-(n-primarybutyl)-p-aminophenol, falling within its scope, then said finding of fact is not necessary. If, however, said claim 28 is construed, but improperly, as a claim, alternative in form, for each of the three compounds, N-(n-primarybutyl)-p-aminophenol, N-(2-ethyl-n-primarybutyl)-p-aminophenol, and N-(2-methyl-n-primarybutyl)-p-aminophenol, then by said Finding of Fact No. 8, it is ruled that there was no invention in providing these latter two compounds. Support for this finding is found in the testimony of Vaughn (Appellant's App. 34) that, with knowledge gained from Bean as to the preparation of N-(n-primarybutyl)-p-aminophenol, it was only a routine matter for him to provide N-(2-ethyl-n-primarybutyl) - p-aminophenol. Where the prior art, by disclosure of N-(n-primarybutyl)-p-aminophenol, so indicated the general line of advance in the field of aminophenols, there could be no invention in providing so closely related a compound as N-(2-ethyl-n-primarybutyl)-p-aminophenol (*L. Sonneborn Sons, Inc.* v. *Coe,* 70 App. D. C. 97, 104 F. (2d) 230; *Sloane* v. *Coe,* 74 App. D. C. 200, 122 F. (2d) 37.

"Conflict of decisions"

7. Under the heading, "Conflict of Decisions," appellant cites two decisions of the District Court, to wit: *Eastman Kodak Company* v. *Coe,* 50 U. S. P. Q. 43 and 645, 40 F. Supp. 891; and *Nye* v. *Coe,* 53 U. S. P. Q. 663, 44 F. Supp. 582. These two cases, it is submitted, are distinguishable on their facts from the case at bar. In the first of these cases, a general formula, wherein certain values might be interpolated, was held inadequate as an anticipation. Here there are several patents specifically describing the compound claimed by appellant. The second of these cases is in the field of machines, rather than chemical compounds, and, except for certain general language, the decision therein has only a remote bearing on the case at bar.

Appellant slights the decision of the Court of Customs and Patent Appeals in *In re Von Bramer and Ruggles, supra.* However, it is plain from the mere language of that decision that that Court ruled adversely to those appellants on many of the contentions raised in this appeal.

CONCLUSION

It is respectfully submitted that the District Court properly followed the decisions of the tribunals of the

Patent Office, and correctly held that claims 28 and 29 on appeal are unpatentable. Therefore, the judgment of the trial court should be affirmed.

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September 1942.

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REPLY BRIEF FOR APPELLANT

IN THE

United States Court of Appeals

FOR THE DISTRICT OF COLUMBIA

Appeal No. 8170

EASTMAN KODAK COMPANY,
Appellant,

vs.

CONWAY P. COE, Commissioner of Patents,
Appellee.

APPEAL FROM THE JUDGMENT OF THE DISTRICT COURT OF THE UNITED STATES FOR THE DISTRICT OF COLUMBIA

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IN THE

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FOR THE DISTRICT OF COLUMBIA

Appeal No. 8170

EASTMAN KODAK COMPANY,

Appellant,

vs.

CONWAY P. COE, Commissioner of Patents,

Appellee.

REPLY BRIEF FOR APPELLANT

1. Reiteration of the Issue

In view of the rambling nature of the Appellee's (Commissioner's) brief, we feel called upon to reiterate that the simple issue involved in this appeal is our submission that the recitation in the prior art of the *mere name* of a chemical compound does not constitute an anticipation of the actual discovery and provision of the compound by Appellant's chemist, Bean, and Bean's full determination and disclosure according to law of the chemical and physical properties of that compound.

At the opening of the Commissioner's brief he occupies four and one-half pages with a complicated discussion of formulae and nomenclature involved in the class of compounds discovered by Bean in an apparent effort to support the idea that prevailed in the mind of the District Court to the very end of the trial of this case that ——

> "I am not talking about the method; I am just saying that you could write that formula and put those chemicals together and get a compound called nitrogen butyl aminophenol." (Page 21 of our main brief)

That is just the point of this appeal—that the mere recitation of a chemical name in the prior art is not evidence of the existence of the compound and is not evidence that the expert chemist——

> "would know that that formula could be gotten together in a crucible, in a chemical laboratory in different ways, with heat applied in different degrees," (page 21 of our main brief)

as stated by the District Court. Not one scintilla of *evidence* of such provision of the compound in the prior art was introduced by the Commissioner at the trial. The District Court's findings were, therefore, not consistent with the evidence because a mere chemical name or formula does not teach the chemist that, like building a playhouse, one merely needs to fit together an OH radical, a benzene ring, an amine group, and a half dozen carbon and hydrogen atoms. On the contrary the provision of the chemical compound requires the discovery of the complicated linkages occurring in that compound, how the intermediates may be obtained and how those intermediates may be finally linked together with the subtraction of certain existing atoms in the intermediates, what catalyst or other conditions may be necessary to cause such linkages to take place and, lastly and possibly most important, the determination and recognition of the various chemical and physical properties of that which is obtained by the foregoing complicated reactions, and proof of the fact that that which is obtained is actually the compound sought.

2. The Prior Art as Evidence

Say what he will, explain what he may, it remains a fact that none of the evidence presented by the Commissioner at the trial shows the existence in such prior art as the Wade British patent, the Clifford U.S. patent and the Rogers and Voorhees application, of Bean's invention.

The Wade British patent discloses *only the name*—"butyl aminophenol."

The Rogers and Voorhees application contains no better disclosure than the Wade patent, it can constitute no better evidence than the Wade patent and the Commissioner so admits.

The Clifford U.S. patent discloses *only the name*—"N-butyl-p-aminophenol" (along with 150 other speculative chemical names) and an inoperative suggestion for making the named compound which Appellant's evidence clearly showed did not produce the compound in question. Merely because Clifford may disclose some other speculative process alleged to produce some *different* compound is no evidence of the compound claimed by Bean. Even though Clifford gives results of alleged tests of N-butyl-p-aminophenol in rubber, it is no evidence of the existence of N-butyl-p-aminophenol inasmuch as Clifford already has failed to give any chemical or physical properties of the compound or any operative process for producing it. Clifford may have tested in rubber, that residue which resulted from carrying out his inoperative process but that does not, *ipso facto,* convert the unknown residue into N-butyl-p-aminophenol.

3. The Incompetence of One Allegation

In addition, this point of rubber tests mentioned in the Clifford patent was not raised by the Commissioner at the trial and the allegation is incompetent at this late stage because Bean has had no opportunity to reply to it and it is quite apparent that it did not constitute a part of the Commissioner's evidence at the trial and no Finding has been made based upon the alleged statement.

4. The Commissioner's Erroneous Approach

Furthermore, the Commissioner has made an erroneous approach upon this appeal. In many places in his brief, Appellee takes the attitude that it is incumbent upon the Appellant to show that the compound of Bean's invention did *not* exist prior to Bean's invention date, that any mere printed statement in the prior art constitutes a fact and that it is incumbent upon Appellant to prove otherwise. Such may be the Commissioner's prerogative in the Patent Office but before a court, legally competent evidence is required. Bean has presented his application to the Patent Office in the manner required by the Rules thereof, fully describing his invention. The Commissioner has refused a patent because of the mere existence of the name butyl aminophenol in the prior art. That does not constitute evidence of the existence of Bean's invention prior to his application and the Findings of the District Court that Bean's invention is anticipated are, therefore, not consistent with the evidence and the District Court should, therefore, be reversed.

5. In re Von Bramer and Ruggles

The appellee's brief urges the applicability of the decision of the U.S. Court of Customs and Patent Appeals in *In re* Von Bramer and Ruggles. The claims in the Von Bramer and Ruggles application were directed to a *motor fuel* and not to a chemical compound as in the instant case. Bean discovered the chemical compounds, Von Bramer and Ruggles sought a patent for a *motor fuel* containing those compounds. The U.S. Court of Customs and Patent Appeals refused a patent to Von Bramer and Ruggles merely because the prior art suggested the incorporation of such types of compounds in motor fuel. In other words, the U.S. Court of Customs and Patent Appeals held there was no invention in such a *motor fuel*. That is a far cry from the issue here involved.

6. Conflict of Decisions

The Commissioner has attempted an answer to the ruling of Justice Bailey in Eastman Kodak Company v. Coe, 50 USPQ 43 but other than to take cover under the *In re* Von Bramer and Ruggles decision, the Commissioner has not weakened the propriety of Justice Bailey's decision.

Only this honorable Court can settle the conflict between Justice O'Donoghue's findings in the instant case and Justice

Bailey's favorable ruling in Eastman Kodak Company v. Coe, 50 USPQ 43. We submit that the principles of law enunciated by Justice Bailey are correct and that the District Court should be reversed in the instant appeal.

Respectfully submitted,

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NEWTON M. PERRINS,
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December 30, 1942.